s

Measuring and Modelling Vehicle NOx Emissions Using a Remote Sensing Device

Christopher Edward Rushton

Institute for Transport Studies

University of Leeds

Submitted in accordance with the requirements for the degree of

Doctor of Philosophy

November 2016

In memory of Robert Worboys

Acknowledgements

I would like to thank everyone who has helped me complete this thesis. My parents who refused to stop supporting me, I could not have done this without them. My supervisors James and Simon for giving me the opportunity to work on this project. The Yorkshire Rams family, especially Mark and Orlando, who taught me how to get the best out of myself. Kev and Yvette for helping me through the final six months. All the people who I have met over the PhD, MSc and undergraduate years who kept me just the right side of crazy and made my time in Leeds unforgettable. Finally my heartfelt thanks go to Volkswagen Group for giving me plenty of research to do over the rest of my career.

Abstract

Despite the increasingly stringent type approval limit values for vehicle emissions no quantitative difference has been seen in roadside concentrations of NO_X concentrations (Carslaw *et al.*, 2011b). This thesis aims to improve the ability of measuring and modelling the NO_X emissions of passenger cars in urban environments by taking an in depth look at the emissions of vehicles observed in real driving environments over a number of years in Aberdeen, Cambridge, Leeds and Sheffield using a Remote Sensing Device. The remote sensing device is tested under controlled conditions to ascertain it's measurement accuracy for both pollutants and vehicle specific power. A mathematical distribution function for describing the emissions of a fleet is presented and shown to be a good description of over 90% of the vehicles and a superposition of two of these distribution functions was able to describe the distribution of the rest of the fleet's NO emissions with a high degree of accuracy. The distribution functions derived for one city were used to create a predictive model to determine how the average emission of a passenger car fleet vehicle performance would evolve over time showing that by 2025 a $\approx$ 30% reduction in NO_X could be expected if the fleet was allowed to evolve naturally.

In addition to these results a number of real world problems were assessed using the new framework developed in this thesis. The emissions of taxis compared to privately owned vehicles was assessed with taxis being shown to emit ≈ 50% more NO_X than their equivalent vehicles in the fleet. The Volkswagen Group scandal, *#dieselgate*, is discussed and the data that the remote sensing device has been used to assess the real driving emissions of VWG passenger cars fitted with the EA189 engine. The observations show that whilst VWG vehicles are exceeding the limit values in real driving environments, they are observed to have equivalent or lower emissions factors than other marques.

Abbreviations

CAA	Clean Air Act
CADC	Common Artemis Drive Cycle
CAN	Controller Area Network
CCM	Corner Cube Mirror
CO	Carbon Monoxide
CO_2	Carbon Dioxide
COMEAP	Committee on the Medical Effects of Air Pollutants
DDIE	Diesel Direct Injection Engine
DOC	Diesel Oxidation Catalyst
DTM	Digital Terrain Model
EC	European Commission
ECU	Engine Control Unit
EPA	Environmental Protection Agency
ETA	1978 Energy Tax Act
EVD	Extreme Value Distribution
FEAT	Fuel Efficiency Automobile Test
FTP-75	Federal Test Procedure 75
GDI	Gasoline Direct Injection
GEV	Generalised Extreme Value
gkm^{-1}	Grams per kilometre
GPS	Global Positioning System
HC	Hydrocarbon
HCV	Heavy Commercial Vehicle
HCK	Hackney Carriage Vehicle
HFID	Heated Flame Ionisation Detector
ITS	Institute for Transport Studies

LAEI	London Atmospheric Emissions Inventory
LEZ	Low Emission Zone
LCV	Light Commercial Vehicle
NAEI	National Atmospheric Emissions Inventory
NEDC	New European Drive Cycle
NO_X	Nitrogen Oxides
NO	Nitric Oxide
NO_2	Nitrogen Dioxide
NDIR	Non-Dispersive Infra Red
NEDC	New European Drive Cycle
N_2	Diatomic Nitrogen
NoV	Notice of Violation
PC	Passenger Car
PEMS	Portable Emission Measurement System
PHEM	Passenger and Heavy Duty Vehicle Emissions Model
PHV	Private Hire Vehicle
RSD	Remote Sensing Device
RDE	Real Driving Environment
RTS	Road Tunnel Study
SAM	Speed and Acceleration Module
SDM	Source Detector Module
TDI	Turbo Direct Injection
TWC	Three Way Catalyst
UDDS	Urban Dynamometer Driving Schedule
ULEZ	Ultra-Low Emission Zone
USA	United States of America
VSP	Vehicle Specific Power
VWG	Volkswagen Group
VOC	Volatile Organic Compounds
WHO	World Health Organisation
WLTP	World Harmonised Light Vehicles Test Procedure

Contents

List of Figures

List of Tables

Chapter 1

Introduction

1.1 Introduction

Air pollution is well known to have significant negative health outcomes when people are exposed to it (COMEAP, 2015; IARC, 2013; Kampa & Castanas, 2008; WHO, 2006, 2013) and costs the economy £16 Billion per year (DeFRA, 2013)[1]. Vehicles have been identified as a major contributor to ambient air pollution concentrations in urban environments (Colvile *et al.*, 2001). Despite legislative interventions to reduce them (EC, 1999a,b,c, 2007b) the emissions by diesel vehicles in urban environments have not decreased as expected (Carslaw *et al.*, 2011a; Williams & Carslaw, 2011). Understanding the reasons there has been a difference between the legally required decrease in emissions and the observed lack thereof, and providing actionable information to help future legislation to be more effective is of critical importance for health outcomes, leading to significant economical savings as well. Steps have been made in identifying trends in vehicle emissions however a deeper understanding of future emissions is required to make informed policy decisions. These decisions have implications beyond the immediate vehicle emissions problem as purchasing behaviour of

---

[1] `https://www.gov.uk/guidance/air-quality-economic-analysis` (accessed 11 February 2016)

vehicle owners will be impacted as well as local economies. It is therefore important that the decisions made relating to vehicle emissions are taken with clear evidence available.

1.2 Background and Motivation

Ambient air pollution in urban environments is responsible for many health problems (Breslow & Goldsmith, 1958) relating to both the cardiovascular system and the respiratory system (Brunekreef, 1997). The result of exposure includes for example a decrease in lung function in children (Gauderman *et al.*, 2004). Recent evidence suggests that exposure to air pollution also leads to a decrease in cognitive ability and provokes depressive-like behaviours(Fonken *et al.*, 2011; Power *et al.*, 2011). These negative outcomes have been shown to be due to exposure to ozone and particulate matter as well as nitrogen dioxide (NO_2) and are present in both children and healthy male adults (Gatto *et al.*, 2014). The contribution by vehicles to this problem is non trivial and has been considered a major factor since 1950. With the increase in petrol powered vehicles with no emission control systems at all came an increase in smog that irritated the eyes. The link between this new pollutant and motor vehicles was proven despite the best attempts of the auto industry to argue that no link existed (De Nevers, 1995). Further research has concluded that there is a significant link between vehicle emissions and a wide range of negative health outcomes (Brunekreef & Holgate, 2002; Janssen *et al.*, 2003; Krivoshto *et al.*, 2008).

There have been a number of significant health events related to high levels of air pollution. In London 1952 over 4000 deaths were attributed to high levels of air pollution lasting 4 to 5 days. Smoke concentrations were measured at between 3 and 10 times their regular levels at all measurement stations and sulphur dioxide levels were between 3 and 12 times their normal measurements at all stations

(Wilkins, 1954). Results from multiple studies have also shown that the extreme values measured in the London event are not the only causes of health problems and that significantly lower air quality concentrations can be just as responsible for negative health outcomes (Schwartz, 1994). It is clear that exposure to air pollution in smaller doses than reported in the most extreme events still presents a danger to the health of the public.

In Europe, as with the rest of the world, there have been numerous legislative attempts to reduce the amount of NO_2 concentrations in urban environments (EC, 2008). The current limit value is $40\mu gm^{-3}$ for annual average concentration, with $200\mu gm^{-3}$ being set as a maximum hourly value. A site may exceed the hourly limit 18 times per year. At some London sites these values were exceeded within one week of the start of 2016 (Guardian, 2016). To reduce the ambient air pollution concentrations regulations have been placed on the emissions of various pollutants from new vehicles. These regulations limit the mass of pollution that a vehicle is allowed to emit over a controlled drive cycle such as the New European Drive Cycle EC (2001) performed in a laboratory (EC, 1999a,b,c, 2007b). The philosophy of this legislation was that by forcing the manufacturers to adopt stricter and stricter limit values it would force them to adapt their technology to become better and better at eliminating poisonous chemicals from their exhaust systems and therefore reduce the concentration of these chemicals in the air.

Evidence suggests that legislation to limit the amount of pollutants emitted by vehicles has not been successful, especially in the case of Diesel vehicles, as they exhibit emission characteristics consistent with far older vehicles operating to less strict emissions limit values (Carslaw & Rhys-Tyler, 2013; Carslaw, 2005; Carslaw *et al.*, 2011b). The current homologation process has been criticised for not accurately representing real world driving and steps have recently been taken to rectify this with the introduction of the Worldwide harmonised Light duty driving Test Cycle (WLTC) (Tutuianu *et al.*, 2013). The WLTC aims to improve the homologation procedure by providing a more realistic drive cycle on which to test vehicles however there are still a number of criticisms. These criticisms relate to the lack of any road gradient variation, which significantly increase engine load and potentially increase the pollution emitted in real world situations. Furthermore the drive cycles themselves are short with the NEDC having a total time of 1180 seconds or less than 20 minutes. These short drive cycles are not representative of the shorter journeys with significant cold start contributions or longer journeys that some vehicles might undertake on a regular basis. Gear shift points are well defined in the NEDC test procedure but are not necessarily representative of real driving behaviour for all drivers. Manufacturers have also been found to program the emission controls to switch off depending on ambient temperature conditions to stop damage to the engine components. There is increasing evidence that manufacturers may be using these loopholes and switching control systems off in milder weather too with the result an increase in polluting chemicals emitted alongside an increase in fuel efficiency (Telegraph, 2016). The increase in fuel efficiency is desirable to manufacturers as it improves the sales potential of the vehicle.

The challenges that these drive cycle limitations place on accurately assessing the real driving environment emissions of a fleet of vehicles means that PEMS, laboratory and drive cycle based tests by themselves are not enough to solve this problem. Remote sensing of these vehicles provides insight that would not be available from other methods. The remote sensing device methodology is, at it's heart, a methodology based in the ideas of big data. A *big data* approach, one where the data is not sampled but is just collected (Mayer-Schönberger & Cukier, 2013), is only currently feasible for vehicle emissions by using a remote sensing device due to limitations of price and vehicle availability (discussed fully in Section 2.5). The big data approach to data processing has been applied in numerous fields most noticeably by (Obama, 2012) in numerous roles across the American government and beyond. Whilst the remote sensing device methodology as presented in this thesis is not as yet a true big data collection methodology as some sampling occurs in terms of site selection it applies the concepts of big data in ways that are not possible in other vehicle emission testing methodologies. Application of a number of the concepts inspired by Mayer-Schönberger & Cukier (2013) to traffic emissions measurements may lead to new insights that would not be found through other more established methods of measurement.

1.3 Aims and Objectives

The primary goal of the thesis is to improve the ability of remote sensing device operators to measure the total oxides of nitrogen ($NO+NO_2$ or NO_X) emissions from vehicles and to improve the ability of policy makers to predict the impact of different policy decisions on the total real driving emissions of vehicles in their fleets. To achieve this a number of methods for analysing remote sensing data have been developed and applied to a number of large data sets collected as parts of collaborations between Aberdeen, Cambridge and Sheffield City Councils. The methods developed have also been used in a series of real world investigations to better understand the behaviour of vehicle fleets in real driving environments.

The first objective, to improve and validate the accuracy of remote sensing currently available NO_X measurements, will take data collected as part of this thesis and investigate the relationships between two different instruments. The efficacy of using data collected by a prototype instrument capable of measuring NO_2 and NO as a basis for estimating total NO_X $(NO + NO_2)$ by the RSD4600, a commercially available instrument with the hardware capability to measure

NO alone, will be assessed. Whilst correlation across instruments has been performed with good results (Bishop *et al.*, 2009) it is currently unknown whether or not the two different instruments produce the same results across a more complete range of vehicles found on the road. Any potential biases are unknown. It is also unknown whether or not using this method will lead to an improvement on the currently used methodologies to calculate total NO_X. The successful correlation between these instruments and successfully showing an improvement on older methods for estimating the NO_2 contribution will allow the best currently available assessment of total NO_X throughout the rest of the thesis and extends to other users of the commercially available RSD4600 equipment.

The second objective is inspired by previous work in the field of astrophysics. Often a simple case of mean and standard deviation are not always appropriate for describing a population. It is believed that a new mathematical framework will provide previously unavailable insights and allow the observations made using remote sensing devices to answer more complex problems relating to the vehicle fleets behaviour. Extreme value theory has been used in pollution concentration studies (Sharma *et al.*, 1999) and attempts have been made to describe vehicle emissions using non-symmetrical distribution functions (Zhang *et al.*, 1994) however the quantity of data along with vehicle metadata currently available means that the opportunity to properly develop this approach using remote sensing data is now feasible. If successful this objective will allow for more naturalistic descriptions of a fleet subset to be constructed using real world data. Deriving a methodology for the application of this branch of mathematics in a meaningful way to vehicle emissions is the first step in being able to create meaningful models that have predictive power as well as simply being able to describe the data already available.

When the analysis of the efficacy of the NO_X measurements has been completed and confirmed the third objective is to create a model that can be used to predict the outcomes of various different traffic interventions in cities where RSD studies have taken place. Not only is this useful for policy decision makers in that city to test the effectiveness of policies, but the lessons learned may be useful to other cities who can extrapolate the impact in their city. In addition to this the production of useful descriptive and predictive methodologies using remote sensing may inspire urban air quality policy decision makers to commission their own remote sensing surveys so as to get a truly bespoke emissions reduction plan.

Given the wealth of remote sensing data currently available from studies conducted throughout the course of the PhD and building on the previously stated objectives, the final objective is to provide insight into the characteristics of smaller subsections of the fleets emissions previously unavailable. Fleet elements such as taxis and light commercial vehicles can readily be investigated using remote sensing device measurements. It is hoped that the robust mathematical description derived previously will provide the tools to achieve this objective. Insights relating to meaningful real world factors such as make, model and vehicle dynamics will help to improve the clarity of the fleet situation and improve the overall results from remote sensing device surveys. The Volkswagen scandal, a developing story throughout the time this thesis was compiled, is the final piece of analysis undertaken.

1.4 Thesis Structure

Chapter 2 provides a literature review covering all the background to the themes investigated in this thesis as well as investigating the steps leading to the current legal framework as that governs both air quality and vehicle emissions. It will assess the effectiveness of these steps and it will also include an overview of techniques that have been used to measure the vehicle contribution to air pollution as well as examining the key air pollution species that will be examined throughout the rest of the thesis.

Chapter 3 will describe the remote sensing methodology. It will discuss the theory, the method of collecting the data, the advantages and disadvantages of the methodology as well as the actual data collected as part of this study. A full description of the sites where data was collected as well as the volume of data collected are also be presented.

Chapter 4 validates the accuracy of the individual modules of the remote sensing instrument. It analyses the accuracy through controlled and repeated measurements performed off the public highway that are atypical of real world remote sensing deployment but provide valuable information about the efficacy of the instrument and approach. This chapter will also present a new method for estimating total NO_X emitted by vehicles using a combination of measurements from two similar but different remote sensing devices, one of which is capable of measuring NO_2 and one which is not. This new methodology, taking observed f_{NO2} measurements and applying them to NO observations, is an improvement over the previous methodology.

Chapter 5 will introduce a new mathematical framework for describing vehicle emissions including their variability using extreme value theory and the Gumbel distribution. It will present a number of real world applications of this methodology using data collected at various sites over the course of the data collection period. It will also present the first steps towards modelling vehicle emissions using the extreme value theory.

Chapter 6 attempts to answer the key research question *how effective has the Euro 6 emission standards been at reducing NO_X emissions?*. It will do this initially by comparing the observed emissions of Euro 6 vehicles to Euro 5 vehicles using the framework developed in Chapter 5 and also by estimating the emissions for ten years using predicted fleet percentages developed by the NAEI in the development of their emission factor toolkit (EFT). The influence of extreme emitters is also discussed and modelled for the same time period to show how effective the introduction of screening based interventions such as stop and check to remove the worst vehicles would be.

Chapter 7 will attempt to answer a series of shorter research questions that the remote sensing data allows us to answer but are not substantial enough to be a chapter by themselves. The effectiveness of the Ultra-Low Emission Zone (ULEZ) proposed for London, Light Commercial Vehicles (LCV's)and Taxis, both private hire and Hackney Carriage style are investigated. The Volkswagen scandal is addressed using data collected using our remote sensing device for the first time and the impact of the newer downsized technology petrol powered vehicles is also assessed.

Chapter 8 will summarise the work done, the impact of the work, the strengths and limitations of the conclusions and look forward to further work and propose additional research questions that could be addressed using remote sensing technology. Final thoughts on the outcomes of this thesis and the future of RSD contributions to the study of vehicle emissions are also presented.

Chapter 2

Literature Review

2.1 Introduction

The following literature review is split into five sections. The first section (Section 2.2) provides a description of the four main pollutants currently considered problematic in the UK. This section will outline both the chemical nature of the molecules and provide an assessment of their behaviour in the urban environment. The health impacts of these pollutants are summarised in both the short term and the long term where appropriate.

The second section (Section 2.3) focuses on the emissions from vehicles. It discusses the sources of the pollutants described in the previous section as well as those pollutants that are not considered in depth throughout the rest of the thesis for context. The section describes the successful use of three way catalysts (TWC's) in reducing the emissions of petrol powered vehicles and shows the chemical pathways that allow them to be successful. The limitations of TWCs are also discussed in that they are not effective on diesel powered vehicles and the chemical and physical processes that can impact their efficiency in petrol powered vehicles. The solutions that have been used in diesel vehicles, considerably more complex than the three way catalyst solution in petrol vehicles and namely Lean NO_X Trap (LNT) Selective

Catalytic Reduction (SCR) and Diesel Particulate Filters (DPF), are described and the limitations of these technologies are explained. The newest technological attempts to reduce emissions to the ever stricter euro standard type approval levels are described and potential problems are also assessed.

The third section (Section 2.4) summarises the legislative steps that have been applied to try and reduce the concentration of pollutants in urban environments and focuses on the legislation that has been introduced to reduce the tailpipe emissions of vehicles. The limit values throughout the history of the implementation of European legislation are shown and the test procedures are described. An assessment of the effectiveness of the current legislative process is provided and a look to the future of limit value legislation is also discussed.

The fourth section (Section 2.5) focuses on alternative methods to the remote sensing device that have been used or are currently in use for measuring vehicle emissions. The methods described are laboratory based measurements, on road tunnel studies, Portable Emissions Measurement Systems (PEMS) studies and vehicle simulation estimates. An assessment of the relative strengths and weaknesses of each method is considered and it is concluded that whilst each measurement method has well defined strengths there are also significant weaknesses with them all. The section concludes that a holistic process involving all of the methodologies is required for a proper assessment of vehicle emissions.

The fifth and final section (Section 2.6) provides a full summary of the history of the remote sensing device methodology that is the primary methodology used for data collection throughout this thesis. The history of the remote sensing device is described from initial concept through to the current commercial equipment used as the primary data collection instrument in this thesis. The prototype instrument that provides additional data used in Chapter 4 is also described. Advantages and limitations of the remote sensing device are described later in Section 3.2.3. A summary of studies performed using remote sensing devices by groups not linked to the Institute for Transport Studies at The University of Leeds (ITS) is provided and a number of studies performed by members closer to the thesis are also summarised.

2.2 Air Pollutants

Air pollutants come in many different forms. This section will describe various air pollutants, both primary and secondary. Primary pollutants that are formed during combustion and in the catalyst process and secondary pollutants that are formed by chemical reactions after they have been emitted from the vehicle. It will examine their sources, their chemistry in combustion and catalytic reactions to remove them from the exhaust plume by after-treatment systems and their interaction with the local environment after they are emitted. The health implications of each pollutant are also summarised. Pollutants such as NO_X which are relevant to this thesis are primary air pollutants along with Volatile Organic Compounds (VOCs), Carbon Monoxide (CO) and Particulate Matter (PM). Secondary air pollutants are formed through the chemical reactions with primary pollutants in the atmosphere after they are emitted from the vehicle. Examples of secondary air pollutants include Ozone (O_3), secondary NO_2 and secondary PM. Whilst background sources such as power stations or factories can influence the ambient concentrations of pollutants in urban environments the source of the majority of primary pollutants in urban areas are vehicle emissions (Bahreini *et al.*, 2012). The emission of primary pollutants in the exhaust plume is is a result of the combustion of either petrol or diesel in internal combustion engines (Colvile *et al.*, 2001) however additional sources of pollutants such as brake disk and pad wear, tyre wear and evaporation can also contribute. This thesis focuses solely on the emission of primary pollutants from combustion. Understanding the sources and behaviour of pollution in urban environments is of high priority because these are the locations where the majority of people are exposed to it and their dose can lead to non-trivial negative health outcomes including but not limited to reduced lifespan.

2.2.1 Particulate Matter

Particulate Matter pollution is commonly referred to in terms of it's cross sectional size in microns ($1 \mu m = 10^{-6} m$). Fine $PM_{2.5}$ refers to a cross sectional size of 2.5 microns and coarse PM_{10} refers to the aerodynamic size, the spherical cross section the particle would have to behave in the observed way, of 10 microns. Ultra fine particles are those with a cross sectional size of 0.1 micron. Particulate matter is produced as part of the combustion process from diesel vehicles and consists of a very complicated combination of particles that exist in the solid, liquid and the gas phase making it very difficult to fully describe and analyse. To further complicate analysis it is possible for liquid particles to adsorb onto solid ones. Secondary particulate matter can form after emission through chemical reactions in the atmosphere. The set of chemicals present is considerable and consists of hundreds of members but contains the notable carcinogens acetaldehyde, arsenic, benzene, dibutyl-phthalate (an endocrine disruptor), formaldehyde, napthalene, benzo-(a)-pyrene and toluene (Wichmann, 2007). A full treatment of the content of the particulate constituent of diesel exhaust emission is beyond the scope of this thesis however in simple terms exposure to the particles emitted by diesel engines has a large number of potential negative health outcomes.

Exposure to increased levels of particulate matter can trigger premature death. The COMEAP (2010) report states that the loss of life expectancy from birth from exposure to anthropogenic particulate matter at 2008 levels in the UK is approximately six months. Alternatively this figure can be expressed as 29×10^3 premature deaths per year. An increase in exposure of $10 \mu g m^{-3}$ for a 24 hour period results in an increase in chances of death of between 0.4% and 1.0% (Pope III & Dockery, 2006).

Links have been drawn between cerebrovascular disease (strokes) in some studies (Hong *et al.*, 2002; Miller *et al.*, 2007) whereas it is not evident in others (Pope *et al.*, 2004). Links have also been drawn between increases in PM exposure and increased risk of deep vein thrombosis (Baccarelli *et al.*, 2008) and cardiac arrests (Forastiere *et al.*, 2005). The totality of the evidence available shows a strong causal relationship between negative cardiovascular health outcomes and exposure to particulate matter pollution with no credible alternative explanation available and as more evidence becomes available it only bolsters the credibility of this argument (Brook *et al.*, 2010). Long term exposure to particulate matter derived from the combustion of fossil fuels has been linked to an increase in the rate of cases of lung cancer (Pope III *et al.*, 2002). Exposure of an additional $10 \mu g m^{-3}$ exposure to $PM_{2.5}$ resulted in an increase in the risk of lung cancer by $14 - 27\%$ in a 26 year study (Turner *et al.*, 2011). As with vascular and pulmonary conditions the currently available evidence suggests that exposure to particulate matter leads to a small increase in mortality and morbidity.

2.2.2 Oxides of Nitrogen

Along with particulate matter, NO_X has been identified as a significant pollutant in an urban environment. NO_X is defined as the total oxides of Nitrogen and generally refers to Nitric Oxide and Nitrogen Dioxide (NO and NO_2). NO is formed by the oxidisation of Nitrogen in the engine combustion chamber and is dependant on the flame temperature (Röpke *et al.*, 1995). NO_X is typically removed from the exhaust gasses of petrol powered vehicles using a Three Way

Catalyst (Section 2.3.2). However in diesel powered vehicles the removal of NO_X is non-trivial. The trade off between NO_X emission and particulate matter emission (Hussain *et al.*, 2012) is one that all engine systems calibrators are aware of. Reduction of oxygen in the cylinder head at the point of combustion via an exhaust gas recirculation system (Section 2.3.4) which also leads to a reduction in flame temperature can reduce the production of NO_X at the expense of PM production. The introduction of a small amount of exhaust gas regeneration can increase the fuel efficiency by reducing the pumping work however exhaust gas regeneration systems can also reduce the fuel mileage and hence increase CO_2 emissions of a vehicle. The final product that is available to buy on the road is therefore a compromise between effective reduction of these three important classes of pollutant.

NO is a primary pollutant produced by mobile sources, through the combustion of both petrol and diesel by internal combustion engine powered vehicles and in static sources such as boilers. It is a colourless gas under standard environmental conditions and contains a free radical unpaired electron (Lund *et al.*, 2011). It is produced in the combustion of hydrocarbons when there is either an excess of oxygen or temperature required for complete combustion of the fuel (Lavoie *et al.*, 1970). Nitric oxide by it's self is less harmful than other primary pollutants however after introduction into the atmosphere Nitric Oxide can undergo a number of chemical reactions leading to secondary pollutants that are considerably less desirable in an urban environment. Secondary NO_2 is produced from NO in the presence of Ozone (O_3). NO and O_3 are produced from NO_2 in the presence of sunlight. Ozone (Section 2.2.3) is a highly oxidising molecule that when inhaled can cause damage to the pulmonary system resulting in significant discomfort and limit exercise performance (Easterly, 1994). Nitric Oxide can also form into nitric acid and secondary particulate pollution.

Nitrogen Dioxide is a soluble reddish brown compound that has strong oxidising capability and has been linked to lung cancer (Hamra *et al.*, 2015). It is produced both as primary emission from oxidation catalysts as fractional NO_2 (f_{NO2}) (Section 2.3.4) and as a secondary pollutant through oxidation of NO in the atmosphere. As an air pollutant NO_2 has numerous roles. Exposure to NO_2 can lead to increased inflammation of airways and increased problems with respiratory function in healthy humans and an increase in respiratory symptoms for those who have asthma (McConnell *et al.*, 2003). Exposure to NO_2 can also lead to a decrease in all-cause life expectancy even when adjustments for particulate matter causes have been taken into account. (COMEAP, 2015) Exposure to concentrations greater than $200\mu gm^{-3}$ are deemed toxic (WHO, 2006). There is significant evidence that exposure to NO_2 can impair the development and growth of lungs (Götschi *et al.*, 2008). It is difficult to detach NO_2 outcomes from the effects of other pollutants that are linked with them such as PM_{10} however the best attempts have been made to decouple the two and show that exposure to NO_2 and the increase in negative respiratory health outcomes are causal however there is not sufficient evidence to confirm that a causal link exists between NO_2 exposure and hospital admissions for cardiovascular problems(COMEAP, 2015).

2.2.3 Ozone

Ozone is not emitted as a primary pollutant by any man-made sources but is formed due to the interaction of other pollutants, namely NO_X and volatile organic compounds found in the particulate matter emissions of vehicles (Defra, 2011). It is a pale blue gas at standard conditions with a distinct pungent smell similar to that of chlorine and detectable in the air at concentrations as low as 10 parts per billion by some people (Hearder, 1874; Rubin, 2001). Ozone is present in the upper atmosphere, stratospheric ozone, in the so called Ozone layer, an area where the concentration of Ozone is approximately 10 parts per million compared to an atmospheric concentration of approximately 0.3 parts per million (McElroy & Fogal, 2008). Ozone in the upper atmosphere is important to life on earth. It is responsible for absorption of the majority of the ultraviolet rays emitted by the sun (Lautenschlager *et al.*, 2007). The absorption of ultra-violet radiation in the upper atmosphere prevents it from reaching the earth's surface and is responsible for preventing incidences of skin cancer (Narayanan *et al.*, 2010).

Ozone at tropospheric levels is far less desirable for humans than stratospheric ozone. Exposure to high concentrations of ozone in humans can result in irritation to the eyes and nose with very high levels of exposure resulting in damaged and inflamed airways, reduced lung function and increased mortality (Defra, 2011). Exposure to ozone concentrations as low as 0.2 parts per million can result in inflammation of the air way and potentially contribute towards asthma and bronchitis (Aris *et al.*, 1993). Children are at an increased risk from exposure to high concentration of ozone. An increase in hospital admissions of 35% for children under 2 years old has been correlated with the increased level of ozone in summer suggesting that the level of ozone in urban environments such as the Toronto area in the study is a risk to young children (Burnett *et al.*, 2001). Exposure to Ozone also potentiates the impact of exposure to particulate matter (Kumarathasan *et al.*, 2015; Vincent *et al.*, 1997). Along with negative health impact on humans high concentration of tropospheric ozone impacts on plant life resulting in significant crop loss (McKee, 1993).

2.2.4 Carbon Monoxide

Carbon monoxide (CO) is a colourless odourless and tasteless gas that is toxic when inhaled by creatures that rely on oxygen transport by haemoglobin. It is produced in incomplete combustion of organic material where there is not enough oxygen present to completely oxidise all of the carbon into carbon dioxide (CO_2). The level of CO at which symptoms of poisoning begin to occur is $35ppm$ with symptoms increasing with greater exposure (Goldstein, 2008). Carbon monoxide poisoning is the greatest cause of injury and death due to poisoning world wide (Thom, 2002). Symptoms of mild carbon monoxide poisoning are headaches, dizziness, drowsiness, headaches, nausea, tachycardia, convulsions, respiratory arrest and death (Goldstein, 2008). It has been shown that exposure to carbon monoxide of children, fetuses and pregnant women can result in an increase in the negative symptoms described previously (Longo, 1977).

2.2.5 Industrial Sources of Air Pollution

The two main sources of air pollution are the transport sector (Colvile *et al.*, 2001) and the generation of electricity with other industrial sources contributing additional smaller amounts of pollution (EEA, 2008). Farming and other regional sources of air pollution contribute alongside global sources both man made and natural in their creation.

Approximately 83% of NO_X in the atmosphere is anthropogenic and the non-anthropogenic component is created by lightning and microscopic organism interactions (Delmas *et al.*, 1997). Mobile transport sources account for approximately half the NO_X emissions with the rest of the anthropogenic component being caused by industrial processes. 40% of the stationary sourced NO_X is produced by electrical power generation plants with the rest being produced by industrial processes that include the combustion of diesel at stationary sources and the burning of biomass (EPA, 1999). Where sources of NO_X are present the secondary pollutant Ozone is also present. Plants can also produce significant amounts of Ozone precursors that can be transformed into Ozone in the presence of sunlight. Given that the overwhelming contributors to NO_X creation, especially in the urban environment are vehicles policies aimed at reducing NO_X concentrations should be focussed primarily on reducing the emissions of NO_X from vehicles.

2.3 Air Pollution in Vehicle Exhausts

2.3.1 The Emission of Air Pollutants

Air pollution can either be emitted into the atmosphere as a primary pollutant or react with other chemicals already present to form a secondary pollutant. As vehicle emissions are the main contributors of pollution in urban environments (Colvile *et al.*, 2001) their characteristics will be the only ones described here. There are four main types of air pollution associated with diesel vehicles. Firstly the emission of unburned hydrocarbons (HC) emissions from diesel engines at low temperature is due to poor fuel air mixture and evaporation and by residual fuel at low load (Boam *et al.*, 1994). Soot or particulate matter (PM) is also produced in diesel vehicles and ranges in diameter from $0.001 - 10\mu m$. Particles with diameter up to $10\mu m$ are defined as PM_{10} and particles with a diameter up to $2.5\mu m$ are defined as $PM_{2.5}$. The ultra-fine fraction of particulates are less than $0.1\mu m$. NO is formed during combustion and can lead to the formation of the secondary pollutants Ozone (O_3) and nitrogen dioxide. Primary nitrogen dioxide (NO_2) is emitted from the tail pipe but is created by the diesel oxidation catalyst (Section 2.3.4) whereas secondary NO_X is created after emission by environmental processes. NO_2 has been shown to have significant negative health impacts (Krivoshto *et al.*, 2008) (Section 2.2.2). Carbon Monoxide (CO) is also produced through incomplete combustion of hydrocarbons due to the lack of available oxygen to fully oxidise the hydrocarbons into CO_2. Carbon monoxide is toxic in high enough concentrations as it is more attractive to oxygen carrying haemoglobin in the blood than oxygen it's self (Anderson *et al.*, 1967).

The driving conditions and driver behaviour as well as the vehicles themselves are responsible for the volume of emissions created with vehicles being the dominant source in urban environments (CMAQ, 2002). Congested conditions are becoming more and more widespread in urban environments (Schrank & Lomax, 2007). Vehicle emissions have been shown to double in links with the same dispersion characteristics from free flow traffic conditions to congested conditions using micro simulation models(Zhang *et al.*, 2011). In studies using PEMS based systems (Section 2.5.2) and experiments based in laboratories (Section2.4.2) short duration acceleration events have been shown to produce particularly high levels of emissions of NO_X in passenger cars (Choudhary & Gokhale, 2016). This is due to increases in engine load (Wenzel *et al.*, 2001). This high level of variability means that modelling these phenomena in a robust way presents a number of difficulties.

2.3.2 Three Way Catalysts in Petrol Engines

The first catalytic converters were introduced in the USA in 1975 however they were only able to remove hydrocarbons and carbon monoxide from the exhaust gasses. These two way catalytic converters were rendered obsolete with the introduction of three way catalytic converters in 1981. Three way catalysts were able to remove NO from the exhaust gas as well as the other two gaseous pollutants. As legislation (Section 2.4) further restricted the levels of emissions required for type approval exhaust gas purification systems have consisted of a catalytic converter and an air/fuel management system.

The aim of the air/fuel management system is to keep the air-to-fuel ratio as close to 14.6 ($\lambda = 1$) as possible. For $1kg$ of fuel burnt $14.6kg$ of air is required for complete combustion. A three-way catalyst, the type typically found on petrol engines, seeks to eliminate Hydrocarbons, Carbon Monoxide and Nitrogen Oxides at the same time by both oxidising the hydrocarbons and Carbon monoxide and reducing the Nitrogen oxides. A catalytic reaction is required to promote these reactions at a lower energy level or temperature than would be required if the reaction was purely thermal. The following reactions represent the ideal oxidisation and reduction processes in a stoichiometric three-way catalytic converter (Heck & Farrauto, 2001).

$$C_yH_{4n} + (1+n)O_2 \rightarrow yCO_2 + 2nH_2O \tag{2.1}$$

$$2CO + O_2 \rightarrow 2CO_2 \tag{2.2}$$

$$CO + H_2O \rightarrow CO_2 + H_2 \tag{2.3}$$

$$NO + CO \rightarrow \frac{1}{2}N_2 + CO_2 \tag{2.4}$$

$$NO + H_2 \rightarrow \frac{1}{2}N_2 + H_2O \tag{2.5}$$

$$(2+\frac{n}{2})NO + C_yH_n \rightarrow (1+\frac{n}{4})N_2 + yCO_2 + \frac{n}{2}H_2O \tag{2.6}$$

While the air to fuel mix remains close to the ratio 14.6 then the three way catalysis process is very efficient. This so called *lambda window* means that the three catalysis processes can happen as efficiently as possible. For air-to-fuel ratios below 14.6 or rich running there are more reducing agents (CO, HC) than oxidising agents (O_2, NO_X) and the NO_X reactions are favoured. Under lean operation the CO and HC reactions are favoured due to the excess of oxygen. By keeping the air to fuel mixture within the lambda window maximal efficiency in conversion of all three pollutants is achieved (Farrauto & Heck, 1999; Lox *et al.*, 1997). Three way catalytic converters store O_2 on the catalyst during periods of leaner burning where it can be released when there is insufficient oxygen to oxidise the hydrocarbon and carbon monoxide components of the exhaust gas (Brandt *et al.*, 2000).

A typical three way catalyst, as used on petrol powered vehicles, consists of a honeycomb monolith support structure covered in a wash-coat, which is then covered in a catalyst material. The honeycomb structure has small channels, typically $1mm$ in diameter. The wash-coat has a thickness of around $20 - 60\mu m$ with a large surface area, approximately $50 - 200m^2g$. The main components used in the wash coats are base-metal oxides such as Aluminium, Cerium and Zirconium alongside Calcium oxide and Magnesium oxide. Rare earth elements such as La_2O_3 (lanthana) are also included to improve the catalytic activity or stability of the wash coat. Cerium has multiple uses. Cerium can be used to store oxygen under lean conditions, hence widening the window where the air fuel ratio yields a value $\lambda = 1$, to stabilise precious metal dispersion and to alter carbon monoxide oxidation kinetics (Lassi, 2003; Lox *et al.*, 1997; Oh & Eickel, 1988). Cerium can also be used to reduce the amount of thermal induced sintering experienced by an alumina wash coat (Gonzalez-Velasco *et al.*, 1994). The precious metals used as catalysts in these systems are Rhodium for NOx and Palladium or Platinum for HC and CO reactions (Armor, 1992; Taylor, 1993; Twigg, 2006). Typical commercial three way catalysts for petrol engines are a combination of two of these metals, for example Pt-Rh (Platinum and Rhodium) or Pd-Rh (Palladium and Rhodium)(Becker & Watson, 1998). It has been suggested that these precious metals or salts thereof may be a cause of health problems if they are emitted from the vehicle as part of the plume. The amounts released are in the order of nano-grams(Artelt *et al.*, 1999; König *et al.*, 1992) however the impact of these particles remains unclear as it is challenging to measure.

The use of three way catalysts at reducing the total emissions of petrol powered vehicles has been very successful. The levels of emissions from petrol vehicles measured in previously conducted remote sensing device studies shows a significant reduction in emissions as successive legislative steps were introduced. The emissions from the most modern vehicles has decreased to the point where their contribution to the overall inventory is considerably less than their diesel counterparts (Carslaw *et al.*, 2011b; Tate, 2016, 2013a,b). There are a number of physical processes that still affect the effectiveness of the three way catalyst such as activation temperature and deterioration of the catalytic material.

Cold Starts and Catalyst Light-Off

The reaction rate of the catalyst is temperature dependent. Light-off is achieved when the conversion rate of exhaust gas components reaches 50% however depending on the test procedure up to 80% of the vehicle emissions over a test cycle can be emitted during this cold start phase. This effect is particularly apparent for CO and HC emissions. Catalyst light off can take up to 2 minutes to achieve (Farrauto & Heck, 1999). Special techniques are employed to decrease the light-off time of the catalyst. These techniques range from passive redesign of the exhaust manifold system to actively increasing the amount of energy in the exhaust gas flow and active pre-heating of the catalyst (Lafyatis *et al.*, 1998).

Moving the pre-catalyst closer to the engine increases the speed at which it absorbs heat from the exhaust gas. Catalyst light off can be achieved in $10s$ in this configuration (Hu & Heck, 1995). Significantly the pre-catalyst in petrol powered engines can be exposed to temperatures in excess of 1000C under certain driving conditions. This high exposure increases the systems vulnerability to sintering and other ageing processes described in Section 2.3.2. The configuration of the pre-catalyst is typically geared towards exothermic oxidation reactions so that the heat can be passed down to the primary catalyst and decrease its light-off time (Becker & Watson, 1998; Lassi, 2003).

The addition of electrical energy to the catalyst monolith can increase the temperature so that light off can occur soon after engine ignition. After reaching the light off point the oxidation reactions can happen and the catalyst can fully heat. Initial designs for electrical heaters required a power demand of in excess of $5kW$ however more modern designs have managed to reduce the power requirement to that available directly from the vehicles alternator (Kubsh, 1994).

The number of vehicles that are emitting at cold start levels rather than optimal levels is unknown. The difference between cold start emissions levels and optimal levels is also unknown. Remote sensing measurements will contain any cold start vehicles that pass through the instrument and may be able to investigate this phenomenon in real driving environments, beginning to quantify the emissions levels produced by these vehicles. Any attempt to quantify this must include accurate data relating to the vehicle temperature and running time. This limitation means that the data used to interpret this must be taken under controlled conditions as the information relating to individual journey time is not available directly from the RSD measurement. Any data needed therefore must be collected independently.

Ageing, Deactivation and Poisoning

The deactivation or ageing of a catalyst is its reduction in conversion efficiency over time due to the removal of effective catalyst sites. This can occur through a number of unwanted physical, thermal or chemical changes within the catalyst monolith structure and attached wash coat. Physical changes can occur through breakages, crushing or general attrition that the system is subjected to through real world driving conditions. The typical drivers of catalyst degradation tend to be associated with thermal and chemical changes in the system. (Carol *et al.*, 1989; Koltsakis & Stamatelos, 1997; Lassi, 2003; Sideris, 1998).

Exposure to extreme thermal energy loading can lead to significant degradation of the catalyst. Whilst a three-way catalyst is designed to be able to withstand short periods of intense thermal energy loading, extended periods of exposure can cause a reduction in efficiency. Exposure to high temperatures for extended periods of time causes reduction in the surface area of the alumina wash coat and sintering of the precious metal catalysts. Sintering of precious metals becomes significant at temperatures in excess of $600°C$ and increases exponentially with temperature increase (Moulijn *et al.*, 2001). The end result is a loss in catalyst surface area which leads to a reduction in conversion of pollutants.

Poisoning is defined as a loss of catalytic activity due to the chemisorption of impurities on the active sites of the catalyst (Lassi, 2003). Poisoning of the catalyst can also occur because of the presence of unwanted chemical components of fuels and lubricants. These impurities are accumulated on the catalysts surface and slowly reduce its efficiency. The difference between poisoning and inhibiting is that whilst poisoning molecules react very strongly and irreversibly with the catalyst sites, inhibitors react weakly and catalyst sites exposed to inhibitors can in some cases be regenerated (Butt, 2012; Butt & Petersen, 1988; Forzatti & Lietti, 1999). Lead, Sulphur, Phosphorus, Zinc, Calcium and Magnesium can all poison catalysts. With the introduction of unleaded fuel, lead poisoning of catalysts, a process that would typically occur after refilling the tank 10 times has become far less significant (Heck & Farrauto, 1996).

Sulphur poisoning still represents a problem to catalyst efficiency and oxygen storage capacity. During the combustion process, Sulphur can oxidise to form SO_2 or SO_3. Sulphur oxides can be adsorbed onto the alumina wash coat at low (less than $300°C$) temperatures, reacting to form aluminium sulphates. This process reduces the alumina surface area of the wash coat and hence the efficiency of the catalyst. At higher temperatures (in excess of 1000°C) sulphur species adsorption is not a significant process (Butt & Petersen, 1988; Heck & Farrauto, 1996). Phosphorus, Zinc, Calcium and Magnesium are often found in lubricant oil and the accumulation of these species on the catalyst surface can cause poisoning of the catalyst (Lassi, 2003). Zinc dialkyldithiophosphate, a common lubricant additive is a common source of both phosphorous and zinc. When combined they can form zinc-pyrophosphate ($Zn_2P_2O_7$). The combination of Zinc and Phosphorous, especially at low temperature can decrease the efficiency of the catalyst more than the presence of each of the individual elements (Williamson *et al.*, 1984). Phosphorus contaminates the catalyst as a layer of Zinc, Calcium and Magnesium phosphates over the catalyst or as Aluminium phosphate within the wash coat (Rokosz *et al.*, 2001).

The choice of catalyst material can also affect the rate of deactivation due to poisoning. Palladium (Pd) is more sensitive to sulphur poisoning than Platinum and Rhodium, for example (Lox *et al.*, 1997). The use of Pd catalysts is only possible now due to the significant reduction in lead content of fuels. Driving conditions are also important. In colder conditions such as North American winters or Nordic conditions, the cooler ambient temperature combined with urban driving conditions can increase the rate of chemical ageing as additional unwanted material is adsorbed onto the catalyst surface (Lassi, 2003).

Three way catalysts have proved to be very effective at removing the three major pollutants, NO, HC and CO. Under ideal operating conditions the efficiency of removing these pollutants can be as high as 90%, 95% and 94% respectively (Hromádko *et al.*, 2010). Limits on CO_2 emissions from vehicles have been introduced to reduce the impact on climate change. To improve marketability of vehicles there has recently been a drive to move away from petrol powered vehicles to vehicles with a lower carbon footprint such as diesel power. These new technologies have their own technical challenges regarding removal of pollutants through catalytic converter reactions.

2.3.3 Alternative Fuels

Compressed natural gas, consisting mainly of methane, is a cheap and abundant fuel with a lower carbon footprint than petrol. This makes it highly desirable as a fuel. However, the high levels of heat and increased amount of poisoning reduce the ability of the three-way catalyst to remove unburned methane more than higher-level alkanes (Winkler *et al.*, 2008). Given that methane has a significantly higher impact as a greenhouse gas than CO_2 (Lashof & Ahuja, 1990) a move to CNG technology should take this into account or it would defeat its own objectives.

2.3.4 Pollutant Removal in Lean Burn Diesel Engines

Lean burn diesel powered engines were designed in part to reduce the carbon footprint but operate in ways that make them unsuitable for three-way catalysts. Increasing engine efficiency through lean burn leads to an increase in NO_X production regardless of the design of the engine (Danaiah *et al.*, 2012). Diesel vehicles are unsuitable for three-way catalysis because the higher level of oxygen in the exhaust gas flow takes the air fuel mix away from the $\lambda = 1$ window discussed in Section 2.3.2. The presence of this oxygen means that effectiveness of reduction reaction of NO_X to N_2 is significantly decreased (Liotta *et al.*, 2002; Russell & Epling, 2011).

Diesel engines have higher fuel efficiency, higher power output per unit mass of fuel and are more durable than petrol engines (Katare *et al.*, 2007). These advantages mean that in principle diesel engines are a highly desirable power unit. The increase in popularity coupled with financial incentives to transition to diesel power mean that the number of diesel vehicles in the fleet has increased in recent years (Haaß & Fuess, 2005). Despite the apparent advantage of diesel engines their fuel burning characteristics mean that they emit more NO than their petrol equivalents. Diesel engines also emit a very complicated particulate matter component in their exhaust plumes containing a volatile organic fraction as well as a solid organic fraction, often with one adsorbed onto the other. Given the short-term health impacts of exposure to diesel fumes and the carcinogenic compounds contained within it, such as NO_2, particulate matter, Benzene and Polycyclic Aromatic Hydrocarbons (PAH) and discussed in Section 2.2, effective control systems are required.

The challenges faced by a system designed to remove pollution from a compression ignition diesel engine are significantly more complex than for a spark, or petrol, powered engine. The high oxygen content of lean burning engine exhaust means that the three way catalyst is unable to function in the same way as in a stoichiometric burning petrol engine. The pollutants dealt with by a three-way catalyst are gas phase whereas the pollutants dealt with by diesel oxidation catalysts are present in all three major phases, solid liquid and gaseous. The emissions from diesel vehicles can also contain complex and volatile organic compounds (VOC's) such as polycyclic aromatic hydrocarbons (PAH's) (Majewski & Khair, 2006). Effectively removing the gas phase pollutants whilst simultaneously removing the solid and liquid phase pollutants is a significant challenge (Farrauto & Voss, 1996). Diesel oxidation catalysts are also unable to significantly self-heat so during periods of low intensity work they may drop below the light-off threshold temperature again (Herreros *et al.*, 2014). The inability of the diesel catalyst to effectively self heat means that in urban driving or congested driving the onboard emission control system is unable to effectively remove the pollution from the exhaust gas unless the engine management system changes the combustion settings and increases the exhaust gas temperature. The inability of the emissions control systems to function effectively in these environments means that the potential for exposure to higher levels of air pollution increase for both drivers in congested traffic and the other users of the urban environment.

There are three main groups of pollution abatement systems each implemented differently depending on the manufacturer. These are Lean NO_X Trap systems (LNT), Exhaust Gas Recirculation system (EGR) and Selective Catalytic Reduction system(SCR). In the Lean NO_X Trap system the first element of the diesel emissions removal system, straight after the exhaust is released from the engine, is the Diesel Oxidation Catalyst (DOC). This component allows CO and NO to be oxidised as well as the hydrocarbons present although it tends not to reduce the amount of total NO_X emitted. The reactions in the DOC also remove the volatile organic fractions adsorbed onto the solid organic soot compounds (Walker, 2004). The oxidation of NO forming NO_2 in the DOC is desirable in theory as it is used in later elements of the emission removal system (Katare & Laing, 2006; Majewski & Khair, 2006) however it is known to be harmful to humans if they are exposed to it (WHO, 2006) Section (2.2). Monitoring of the trends of fractional NO_2 emissions suggest that it is increasing (Carslaw, 2005).

After the DOC a Lean NO_X Trap (LNT) or NO_X Storage and Removal (NSR) module can be introduced. These modules perform better when there is more NO_2 present in the exhaust gas (Epling *et al.*, 2004; Olsson & Fridell, 2002; Olsson *et al.*, 2001; Prinetto *et al.*, 2001; Twigg, 2006). An NSR catalyst aims to store the NOx as a nitrate under lean burn conditions and periodically remove it under rich burn conditions managed by the engine control unit (Matsumoto *et al.*, 2000). During lean burn periods the NO is oxidised over a Platinum catalyst to create NO_2. The resultant NO_2 is captured by catalyst compounds containing barium, for example barium Oxide (BaO) is often used alongside platinum (Pt), to create platinum oxide. During the short rich burning periods of operation, the NO_X is released from the barium and reacts with hydrocarbons to produce N_2, H_2O and CO_2 (Olsson & Fridell, 2002).

LNT / NSR catalyst systems are vulnerable to poisoning from Sulphur. The Sulphur can react with both the precious metal catalyst elements as described before and with the NOx storage elements such as Barium to form Barium sulphate ($BaSO_4$). Introducing a combination of Titanium and Lithium doped Al_2O_3 to the catalyst can mitigate this effect (Matsumoto *et al.*, 2000). Sulphur poisoning can also be removed by exposure to high temperatures removing the sulphur compounds and allowing the barium to adsorb NO_X (Epling *et al.*, 2004).

The final part of modern diesel exhaust after-treatment systems are Selective Catalytic Reduction systems (SCR). SCR are able to reduce the amount of NO_X present in vehicle emissions by the introduction of an additional compound post combustion. Whilst SCR catalysts using platinum, iridium and silver are possible the most common SCR system uses ammonia (NH_3) over a vandium, copper or iron catalyst (Gieshoff *et al.*, 2000; Girard *et al.*, 2009; Russell & Epling, 2011). There are three SCR reactions possible to reduce NO_X. The standard reaction is shown in Equation 2.7, the fast reaction in Equation 2.8 and the NO_2 only reaction in Equation 2.9. When there is an equimolar mixture of NO and NO_2 the fast reaction takes place leading to improved performance at lower temperatures (Nova *et al.*, 2006).

$$4NH_3 + 4NO + O_2 \rightarrow 4N_2 + 6H_2O \tag{2.7}$$

$$4NH_3 + 2NO + 2NO_2 \rightarrow 4N_2 + 6H_2O \tag{2.8}$$

$$8NH_3 + 6NO_2 \rightarrow 7N_2 + 12H_2O \tag{2.9}$$

The SCR reaction, unlike the previous reduction reactions used in the three way catalyst, are not affected by the presence of oxygen and are ideal for use in lean burn diesel engines. The addition of the SCR catalyst means that an ammonia slip catalyst is also required as ammonia can cause health problems if inhaled (Carson *et al.*, 1981). The efficiency of SCR reactions are dependent on both the ammonia dose and the gas temperature. With a gas temperature of $360°C$ a ratio of ≈ 1.1 $NO_X : NH_3$ is required, rising to ≈ 1.3 for a temperature of $420°C$. At lower temperature the percentage of ammonia slip, lost from the tailpipe is higher than at higher temperature for the same $NO_X : NH_3$ ratio (DieselNet, 2005). Fine tuning of the SCR system can achieve very good reduction of total NO_X emissions (Carslaw *et al.*, 2015).

Exhaust Gas Recirculation systems operate by re-introducing some of the exhaust gas back into the cylinder head. This serves to reduce the peak temperature in the cylinder head and therefore reduce the total NO_X that is created through combustion (Section 2.2.2). This process works because the exhaust gas has a higher specific heat capacity than air but as a result reduces the maximum power that can be generated. The drawbacks of the EGR systems also include dirtying of the engine leading to a decrease in performance an increase in soot production which leads to diesel particulate filters being installed which can increase back pressure and reduce performance (Dagel, 1998).

Diesel Particulate Filters (DPFs) are the final piece of the diesel engine emission control system and often located after the DOC. A DPF is most commonly a wall flow monolithic structure with cells extruded axially and plugged at alternate ends forcing the exhaust gas to pass through the cell walls where the particulate matter is adsorbed onto the surface. DPF systems are most effective at removing the solid fraction such as elemental carbon or soot from the exhaust plume however they are not effective at removing the liquid fraction. All DPF systems that are relevant to removal of the PM element from diesel exhaust gasses are based on the concept of a particle trap as the low density and small size means that gravitational or centrifugal based filters are ineffective as they behave more like a gas than a solid in a cyclone. Introduction of an electrostatic element to the cyclone system would improve the efficiency of the system however they appear to be impractical when applied to diesel engines (Majewski, 2001)

Buildup of soot or Particulate Matter (PM) can increase the back pressure and reduce the efficiency of the engine or cause a complete failure. PM can be reduced but not completely eliminated from the exhaust by modifications to the engine design (Twigg, 2006) and a DOC can oxidise hydrocarbons of the Soluble Organic Fraction (SOF) of PM to further reduce the amount of soot (Russell & Epling, 2011). These deposits are removed through oxidation to other gasses, primarily CO_2, in a process called thermal regeneration. In passenger vehicles periodic regeneration of the DPF is required to remove the carbon elements that are adsorbed onto it. This is achieved by increasing the exhaust temperatures to PM oxidising temperatures of approximately $500°C$, by injecting fuel that is burned over a Pt catalyst either as part of the DOC or upstream of it (Russell & Epling, 2011; Watanabe *et al.*, 2007). The exhaust temperatures of light duty diesel engines are in the range of $100°C$ to $200°C$. The required increase in exhaust gas temperature can be achieved in a number of active and passive methods including fuel-born catalysts, catalytic filter coatings and generation of reactive species such as NO_2 (Hawker *et al.*, 1997) along with active methods such as fuel burners, electric heating, microwave heating and the injection of fuel into the exhaust gas (Konstandopoulos *et al.*, 2000).

In real life the DPFs in commercial vehicles are regenerated by burning the soot from the filter. This can occur either after extended periods of high engine load resulting in high exhaust temperature such as occurs in extended motorway driving or hill climbing or can be forced when the filter reaches a given capacity. The forced regeneration is induced by the introduction of fuel post combustion and can occur in any situation and in drive cycle tests it is often observed after the extra-urban cycle in the urban cycle (EA, 2013).

The system required to remove pollutants from diesel exhaust is complicated with many individual chemical reactions required to produce the desired clean air. Modern ECU's give engineers great control over the behaviour of their engines however when measured on the road the evidence shows that these complicated systems whilst passing the required emissions type approval tests are not successful at reducing the emissions of vehicles in real driving environments (Carslaw *et al.*, 2011a,b). A greater understanding of the on-road sources of these emissions can only help to eventually reduce the total emissions produced and in turn the roadside concentrations and hence the negative health impacts caused by these pollutants.

2.4 Air Quality Legislation

2.4.1 Summary of Legislation

The impact on health due to air pollutants has been discussed in Section **??**. To limit the negative health outcomes experienced a number of limit values have been introduced for urban environments. For NO_2 the limit value is $200 \mu g m^{-3}$ averaged hourly with 18 permitted exceptions per year and an overall annual average of $40 \mu g m^{-3}$. Limit values for the annual average concentration of particulate matter with a cross section of 2.5×10^{-6} and 10×10^{-6} ($PM_{2.5}$ and PM_{10}) are

$25\mu gm^{-3}$ for $PM_{2.5}$ and $40\mu gm^{-3}$ for PM_{10}. PM_{10} has a daily concentration limit value of $50\mu gm^{-3}$ with 35 exceptions allowed. The limit value for CO is $10\mu gm^{-3}$ for the maximum daily 8 hour mean and the limit value for O_3 is $120\mu gm^{-3}$ for the maximum daily 8 hour mean. The limits on hydrocarbons are specific to different species (EC, 2008).

The majority of these limit value exceptions occur in urban environments close to roads and are likely to be caused by diesel vehicles on top of background sources (Carslaw *et al.*, 2001). To reduce the negative health outcomes related to poor air quality various steps have been taken to limit the NO_X emissions from new vehicles vehicles. These legislative steps have taken the form of limit values on a vehicles emission factor expressed in grams per kilometre (gkm^{-1}) averaged over a chassis dynamometer drive cycle and are described in Section 2.4.4. There are a number of recognised drive cycles currently in the literature and their advantages and disadvantages are summarised in Section 2.4.3.

2.4.2 Laboratory Based Dynamometer Tests

Vehicle emissions measurements have typically been made using dynamometer based tests under controlled laboratory conditions and the type approval tests are always performed on them. A dynamometer is a device that is used to measure force (from the CGS force unit Dyne $1dyn = 10^{-5}N$). A brake dynamometer applies variable load to a driven element and calculates the torque required to drive it and given the term rolling road. An inertial dynamometer uses an element of fixed inertia and calculates the torque required to rotate it. Both types of dynamometer can be used to perform sweep tests to analyse an engines power output and emissions generated. Dynamometer tests are of two types: Engine Dynamometer Tests and Chasis Dynamometer Tests. In Engine Dynamometer Tests the engine is fitted to a test rig and then directly to a dynamometer and in Chassis Dynamometer Tests the whole vehicle is tested on a rolling road capable of providing dynamometer style responses creating resistance calculated from coast down tests (Eastwood, 2000). The laboratory provides great opportunity to test vehicle emissions in depth with a wide range of equipment. Heated Flame Ionisation Detector (HFID) analysis, Non-Dispersive Infra Red (NDIR) chemo-luminescence analysis, electro-chemical analysis and sample traps have been reported (Turrio-Baldassarri *et al.*, 2004). Further analysis of the speciation of vehicle emission including the Volatile Organic Compounds (VOC's) is also possible using real-time chemical ionisation mass spectrometry (Heeb *et al.*, 2003).

The main advantage of lab based tests is the standardisation. Strict operating conditions and very precisely calibrated equipment mean that the procedures produce highly repeatable results that are well suited to the creation of baselines and comparative tests. For type approval tests the initial conditions and the drive cycle are very well defined (EC, 1999b, 2007b) and described in 2.4.3. Generally highly repeatable measurements are desirable in a scientific context however these laboratory based tests do not account for the real driving environment that vehicles are expected to perform in on a day to day basis. Vehicle dynamics on standard drive cycles such as the New European Drive Cycle (NEDC) (EC, 2001) are not representative of the power demands experienced by the engine in the real world. The relationship between engine load has been shown to be significant and non-linear with disproportionately high emissions observed for higher power requirements (Andrews & Ahamed, 1999). In locations where real driving features a high fraction of off cycle conditions laboratory derived results are likely to lead to an underestimate the total vehicle emissions. Furthermore after the Volkswagen group diesel engine scandal it has been shown that the standardisation of laboratory tests can be exploited to trigger different emissions control system strategies in the ECU.

A further criticism of the laboratory based tests is the vehicles tested. New or well maintained vehicles are typically chosen for tests and these so called *golden vehicles*, those which perform particularly well, are not necessarily representative of all vehicles on the road. Vehicle maintenance has been shown to be an impacting factor in that vehicle's emissions with some studies suggesting that 10% of the vehicles being responsible for 50% of the emissions (Bishop *et al.*, 1997; Zhang *et al.*, 1996b). The failure of laboratories to account for these high-emitting vehicles can lead to a significant distortion of the total calculated emissions (Ropkins *et al.*, 2009).

The criticism levelled at laboratory tests in this section should not be taken as evidence that the methodology does not have it's uses. Laboratories provide significant advantages over on-road systems in terms of instrumentation however their limitations should also be noted. The laboratory method has a number of key failings that mean that they are unsuitable for measurements related to real driving emissions. A holistic approach that combines on-road and off-road measurements should be used if a highly accurate picture of vehicle emissions is to be created.

2.4.3 Drive Cycle Description and Analysis

New European Drive Cycle

The NEDC (EC, 2001) consists of four identical urban drive cycles and one extreme urban drive cycle and lasts 1180 seconds. The test procedure may be conducted on a road but for repeatability and consistency purposes it is typically conducted in a laboratory on a dynamometer. The vehicle to be tested must be a cold start and at a temperature of between $20°C$ and $30°C$ after a heat soak of at least

6 hours and allowed to idle for 40 seconds. All ancillary systems such as air conditioning, heated windows and lights are switched off.

The NEDC has attracted criticism for a number of reasons. The drive cycle it's self although being described as an urban drive cycle shows no resemblance what so ever to a vehicle being driven in an urban environment by any real world driver. Power demands are significantly different to real world drive cycles with regards to acceleration Andrews *et al.* (2004, 2005); Samuel *et al.* (2004). Vehicle Specific Power (VSP) Jimenez-Palacios (1998) measurements derived from on-road measurements have also shown to diverge from the range tested under the Common Artemis Drive Cycle André (2004) (CADC) Chen & Borken-Kleefeld (2014). The initial temperature conditions specified for the vehicle are unrepresentative of the real world in the vast majority of cases. Temperatures of 20°C are not representative of the UK where the maximum average temperature by month in London is 19°C [1]. Ancillary systems such as air conditioning fans and pumps require significant amounts of electrical energy to function and switching them off significantly reduces the load required on the engine. For example a 200W electrical load can reduce engine fuel efficiency by $0.4 Lkm^{-1}$ ($0.94 mpg$) (Kassakian *et al.*, 1996). The removal of these systems from the test procedure is unrepresentative of real world driving especially if the ambient temperature is in excess of 20°C.

Common ARTEMIS Drive Cycle

The Common ARTEMIS Drive Cycle (CADC) is a chassis dynamometer drive cycle developed by the European Assessment and Reliability of Transport Emission Models and Inventory Systems (ARTEMIS) project. It was developed using a data acquired from 58 real vehicles driven for their usual purposes creating a database of $73000 km$, 8200

---

[1] http://www.holiday-weather.com/london/averages/

trips and 1680 hours of real driving. The data collected was then used to generate a quasi-realistic drive cycle (André, 2004).

The CADC was originally designed to be a more realistic and hence dynamic cycle and hence improvement on the NEDC that could be used as part of the ARTEMIS framework and also in external projects that were attempting to reduce vehicle emissions (André, 2004). Analysis of real world vehicle engine demands shows that the CADC is still not capturing the full range of vehicle dynamics that are exhibited in real driving conditions (Chen & Borken-Kleefeld, 2014) but is more representative of real driving than the NEDC.

Federal Test Procedure 75

The Federal Test Procedure 75 (FTP-75) is the equivalent to the NEDC in the USA measuring tailpipe emissions and CO_2 emissions. It was initially introduced as part of the 1978 Energy Tax Act (ETA) to determine the rate of the guzzler tax to be applied to new cars. It was last updated in 2008 and includes four phases. The urban cycle (FTP-75 proper), highway cycle (HWFET), aggressive cycle (SFTP US06) and an optional air conditioning cycle (SFTP SC03). A similar but truncated version known as Urban Dynamometer Driving Schedule (UDDS) also known as FTP-72 is used in Sweden and Australia as their vehicle emissions drive cycles. The NO_X limit values for Tier 2 vehicles such as those tested in the VWG scandal is $0.043gkm^1$ compared to the $0.18gkm^1$ Euro 5 and $0.08gkm^1$ Euro 6 limit values.

The FTP-75 proper cycle is one cold start run of the FTP-72 followed by a 10 minute period where the engine is switched off and finished with the first 505 seconds of the FTP-72 cycle repeated from a hot start. The total distance covered is $17.77km$ at an average speed of $34.1kmh^1$ and lasts for 1874 seconds. The weighting between the hot and cold starts are $1 : 0.43$ respectively.

The highway driving cycle (HWFET) is a 765 second cycle with no stops using a warmed up engine. It has an average speed of $77.7kmh^1$ and a maximum speed of $97kmh^1$. The SFTP-US06 element was introduced to address the shortcomings of the FTP-75 cycle and has extended periods of aggressive driving more representative of real world conditions. It lasts 10 minutes, covers 13km, averages $77kmh1$ and has a top speed of $130kmh^1$. The maximum acceleration is $13.6kmh^{-1}s^{-1}$ ($0.756ms^{-2}$). A warm start engine is used, air conditioning is switched off and ambient temperatures are between 20°C and 30°C. The optional air conditioning drive cycle is 596 seconds long, length 5.8km with an average speed of $34.8kmh^1$, a maximum speed of $88.2kmh^1$. Ambient temperatures are increased to 35°C and the engine is started warm.

World Harmonised Light Vehicle Test Procedure

The WLTP is part of the initiative to harmonise vehicle emission standards world-wide. It is currently under development by teams from the European Union, India and Japan and finalised versions were expected in October 2015. The test procedure splits vehicles into one of three classes depending on their power to weight ratio with the majority of modern European vehicles belonging to Class 3 (power to weight ratio $> 34kWT^{-1}$) although some busses may belong to Class 2 (power to weight ratio $> 22kWT^{-1}$ and $\leqslant 34kWT^{-1}$).

One major criticism of the NEDC is that it fails to represent real world driving and hence real world NO_X emissions. In deriving the WLTP research has been completed to identify where in the speed and acceleration solution space the majority of the NO_X is emitted (Demuynck *et al.*, 2012). By applying this knowledge to the WLTP it should become harder to cheat the test. There still remains a shortcoming in the $70kmh^{-1}$ to $110kmh^{-1}$ speed bracket which could potentially be an area for concern for some vehicles Sileghem *et al.* (2014). A further criticism of the NEDC procedure is the ambient temperature requirements. Whilst significantly lower than the 1000°C temperatures some catalysts can be exposed to during driving, the 20°C−30°C temperature requirements of the test are far higher than would normally be expected. It has also been shown that the WLTP test procedure needs to robustly take account of the cold start procedures as the initial phase of the test can significantly impact the total NO_X emissions from the test Sileghem *et al.* (2014).

Despite being an improvement over the NEDC criticisms of the WLTP still exist. It is generally quite slow with the quickest $0 - 50kmh^{-1}$ time being $15s$, slower than the typical $5 - 10s$ range expected for normal driving. It also fails to include road gradient. Road gradient has been shown to have a significant effect on engine power demand and hence vehicle pollution (Wyatt *et al.*, 2014). There is also an issue in the $70kmh^{-1}$ to $110kmh^{-1}$ speed bracket of the speed and acceleration solution space which could potentially be an area for concern for some vehicles based on their emissions over the CADC as their total emissions would be underestimated Sileghem *et al.* (2014).

Relevance to Real Driving

Compared to the NEDC the FTP-75 along with the supplemental tests is far more representative of real world driving however the current regulations are set to be superseded by the World Harmonised Light Vehicles Test Procedure (WLTP). A comparison of the Vehicle Specific Power (Jimenez-Palacios, 1998) between the NEDC, the FTP-75 and the RSD measurements taken in both Cambridge (Tate (2013a), Section 3.4.2) and Sheffield (Tate (2013b), Section 3.4.1) and displayed as a data density plot alongside a drivecycle timeseries in Figure 2.1 show that the engine power requirements that a vehicle is subjected to (VSP, Section 4.2.1 Equation 4.1) whilst taking the type approval test are not relevant for real driving in urban environments. The Cambridge and the Sheffield site locations cover a wide range of

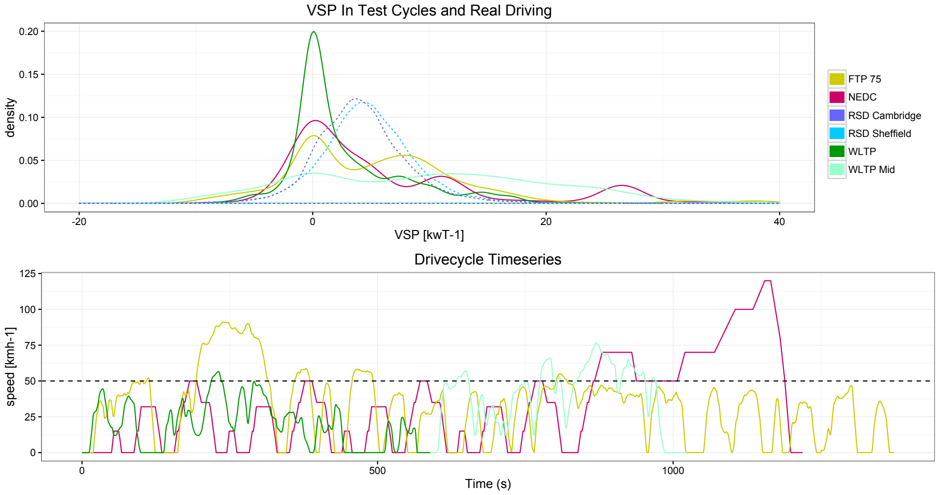

Figure 2.1: Vehicle specific power demands across different drive cycles

real driving environments and gradients and are broadly representative of real conditions when considered in totality. The time series plot shows that rarely under either the NEDC or the WLTP cycles does the $30mh^{-1}$ $(50kmh6-1)$ speed limit get reached or broken. This again is highly unrepresentative of real driving.

Apart from a small number of VSP measurements on the NEDC the drive cycle is not representative in terms of the power demands experienced by vehicles driving in real environments. The small number of occasions where the VSP is relevant to real driving demands is still lower than the modal number for both urban environments observed. Despite an increased number of higher VSP demand intervals the WLTP low speed cycle, the section that represents urban driving is still not representative of the RSD data. The medium speed section of the WLTP drive cycle does include higher power engine demands however it is not supposed to take into account urban driving. Concerns still remain about the ability of drive cycle tests to accurately represent real driving conditions. Remote sensing device measurements are biased towards areas of higher VSP as it improves the measurement success rate (Section 3.2.3) however NEDC does not provide any coverage of the VSP demanded at RSD sites. The WLTP begins to take this into account however.

2.4.4 Vehicle Emission Legislation

New vehicles that seek type approval after 2014 will have to meet Euro 6 emissions (EC, 2007a) which follow the previous trends of reducing vehicle emissions but currently still use the NEDC. Initial testing using Portable Emission Measurement System methodologies have shown that a Euro 6 vehicle fitted with a Selective Catalytic Reduction (SCR) system emits less NO_X than a Euro 5 or Euro 4 vehicle but is still significantly over the limit for type approval (Weiss *et al.*, 2012). A vehicle must undergo this type approval process before it an be sold in the EU. The EU directives related to vehicle emission limit values expressed in grams per kilometre are summarised in Table 2.4.4 and Table 2.4.4 (EC, 1991, 1994, 1996, 1999a,b,c, 2007a,b). Prior to September 2017 all of the limit values presented must be achieved over the NEDC however after September 2017 the regulations change and new vehicles must achieve these limit values over the WLTP (Section 2.4.3) as well as over a 90 - 120 minute real world driving test measured using a PEMS device (Section 2.5.2).

The Euro 6 legislation introduced more stringent testing requirements for vehicles in both limit values and testing methodology. Euro 6a vehicles were subject to the same tests as Euro 5 albeit with reduced emissions factors. Euro 6b vehicles which will be introduced in 2018 for new type approvals will require that the vehicle meet the same limits but over the WLTP test cycle (Section 2.4.3). Euro 6c vehicles will be required to meet these tests using PEMS (Section 2.5.2) measurements to calculate real driving emission (RDE) subject to a conformity factor (CF), a fraction by which the real driving performance is allowed to exceed the NO_X limits, the value of which remains

Tier	Type Approval Date	CO	HC	NO_X	PM
Diesel Car					
Euro 1	July 1992	2.72	-	-	0.14
Euro 2	Jan 1996	1.0	-	-	0.08
Euro 3	Jan 2000	0.64	-	0.5	0.05
Euro 4	Jan 2005	0.5	-	0.25	0.025
Euro 5	Sept 2009	0.5	-	0.18	0.005
Euro 6	Sept 2014	0.5	-	0.08	0.005
Petrol Car					
Euro 1	July 1992	2.72	-	-	-
Euro 2	Jan 1996	2.2	-	-	-
Euro 3	Jan 2000	2.3	0.2	0.15	-
Euro 4	Jan 2005	1	0.1	0.08	-
Euro 5	Sept 2009	1	0.1	0.06	0.005
Euro 6	Sept 2014	1	0.1	0.06	0.005

Tier	Date	CO	HC	NO_X	PM
LCV NI					
Euro 1	Oct 1994	5.17	-	-	0.14
Euro 2	Jan 1998	1.25	-	-	0.08
Euro 3	Jan 2000	0.8	-	0.50	0.05
Euro 4	Jan 2005	0.63	-	0.25	0.025
Euro 5	Sept 2009	0.63	-	0.18	0.005
Euro 6	Sept 2014	0.63	-	0.105	0.005
LCV NII					
Euro 1	Oct 1994	5.17	-	-	0.19
Euro 2	Jan 1998	1.25	-	-	0.12
Euro 3	Jan 2001	0.8	-	0.65	0.07
Euro 4	Jan 2006	0.63	-	0.33	0.04
Euro 5	Sept 2010	0.63	-	0.235	0.005
Euro 6	Sept 2015	0.63	-	0.105	0.005
LCV NIII					
Euro 1	Oct 1994	6.9	-	-	0.14
Euro 2	Jan 1998	1.5	-	-	0.08
Euro 3	Jan 2001	0.95	-	0.78	0.05
Euro 4	Jan 2006	0.74	-	0.39	0.025
Euro 5	Sept 2010	0.74	-	0.28	0.005
Euro 6	Sept 2015	0.74	-	0.125	0.005

an open topic of debate but is currently set at the values defined in Table 2.4.4.

Weiss *et al.* (2012) compared a Euro 6 diesel car fitted with an SCR system to a set of six Euro 4 and 5 diesel cars and found a reduction in NO_X emission of a factor of 4. Andersson *et al.* (2014) analysed 2 diesel euro 6 vehicles using both PEMS and drive cycle analysis and found that whilst both vehicles passed the Euro 6 limit value on the NEDC cycle when other cycles or PEMS testing was used the emissions were higher than for the NEDC and exceeded the Euro 6 limit value. Yang *et al.* (2015b) further extended this analysis by studying 73 Euro 6 diesel vehicles in laboratory settings using the NEDC and the WLTP cycle. All the vehicles tested in this scenario performed well on the NEDC but were poor on the WLTP cycle. Furthermore five vehicles fitted with Lean NO_X Traps (LNTs) were observed to emit significantly higher emissions than the rest of the Euro 6 fleet over the WLTP. These results were also observed in the ICCT white paper (Yang *et al.*, 2015a) where 3 LNT equipped Euro 6 diesel cars emitted and in a further ICCT report (Franco *et al.*, 2014) where both a vehicle fitted with LNT and one with SCR systems were emitting much higher than the rest of the vehicles tested. O'Driscoll *et al.* (2016) tested 39 Euro 6 vehicles with a range of LNT, EGR and SCR systems using PEMS. They showed that two vehicles met the Euro 6 limit and a further two were within 10% of it. A LNT vehicle and an SCR / EGR vehicle were able to pass the limit values in real driving showing that it is possible to achieve these values. Of the 37 vehicles that failed 22 vehicles failed the limit value with their NO_2 emissions alone and average fractional NO_2 values were $44\% \pm 20\%$ although the range of observations is not consistent across any subset of vehicles. The mean NO_X emissions from these vehicles would require a conformity factor of 5.3 for urban and 3.1 for motorway driving compared to a target conformity of 2.1 for new vehicles in 2019 falling to 1.5 in 2021 (EC, 2015). A remote sensing device can test

Tier	New Registration Date	NO_X	Requirements
Euro 6a	Sept 2014	0.08	NEDC
Euro 6b	Sept 2018	0.08	WLTP
Euro 6c	Jan 2019	0.08	RDE CF - 2.1
Euro 6c	Jan 2021	0.08	RDE CF - 1.5

a large number of Euro 6b / 6c vehicles and assess how they are performing relative to one another and to the required limit value and conformity factors.

Along with type approval limit values other legislative steps have been introduced to reduce the concentration of pollution in urban environments. The London congestion charge was introduced in 2003 to reduce the number of vehicles in London centre and also as a way to limit the concentration of pollution in the city centre with reductions as high as 30% predicted (Daniel & Bekka, 2000). A number of low emission zones (LEZ's) have been introduced across Europe to attempt to further reduce vehicle emissions by only letting vehicles that meet the low type approval values for air pollution emissions required by more modern vehicles into designated zones. Approximately 200 LEZ's have been created around Europe with the UK having 3, Germany approximately 70 and Italy approximately 90. LEZ's typically restrict the access of Heavy Commercial Vehicles (HCV's) although they vary from zone to zone with restrictions being placed on Light Commercial Vehicles (LCV's) and cars in some examples (Holman *et al.*, 2015).

2.4.5 Effectiveness of Legislative Steps

The ultimate test of the effectiveness of these legislative steps is an observed decrease in concentration of pollutants in urban environments. Such success has been observed for HC and CO with the introduction of a TWC (Mayer, 1999). Carslaw *et al.* (2011a) measured ambient NO_X and NO_2 concentrations at 39 sites across the UK. 11 roadside sites, 11 urban centre sites and 17 urban background sites. Furthermore in London 10 inner London, 13 outer London and 19 London background sites were measured up until the end of 2008. Trends in NO_X and NO_2 since 1996 can be categorised into 2 phases. From 1996 to 2004 a general decline in NO_X and NO_2 was observed across all sites. This period of decline was followed by a period of stability at best described as a period of slow decline. NO_X concentrations for UK urban centres changed by -0.8% compared to a change of -0.6% in inner London. The general trend of a period of decline followed by a period of stability is observed across Europe with variation on the trend observed in some countries such as Greece where a much larger initial decline is observed in the 1996 to 2004 period.

The efficacy of the London congestion charge system at reducing air pollution concentrations was initially assessed at -12% in the urban environment (Beevers & Carslaw, 2005) although there was no noticeable change in background NO_X concentration (Atkinson *et al.*, 2009). The London LEZ began operation in 2008 and at 1500 at $1500km^2$ is the largest in the world and reports a 98.9% compliance (Holman *et al.*, 2015). The decrease in NO_X was estimated at -20% in central London and -19% in inner London (Cloke *et al.*, 2000) however no discernible impact has been observed in the NO_X concentrations and only a small decrease has been observed for particulate matter (Ellison *et al.*, 2013). Generally it has been difficult to show that there has been any significant reduction in NO_X or PM_{10} resultant from the introduction of Low Emission Zones (Holman *et al.*, 2015). The reason behind the lack of any change in ambient pollution is most likely due to the fact that vehicle NO_X emissions in real driving environments has not decreased in line with the required limit values (Carslaw & Rhys-Tyler, 2013).

Motivated by the general failure of the London LEZ to meet any of it's stated aims additional regulations have been put in place to attempt to reduce the ambient concentrations of NO_X. From 2020 an Ultra Low Emission Zone (ULEZ) will be introduced into London covering the same area as the congestion charge. In this area all vehicles will have to be compliant 24 hours a day 7 days a week. For diesel passenger cars and light commercial vehicles this means that only those that meet the Euro 6 regulation will be allowed into the zone without having to pay a charge of £12.50 per day. Petrol passenger vehicles and LCVs that meet the Euro 4 type approval limit values will also be permitted. Busses and HCVs will be required to meet the Euro VI standard and motorcycles will have to meet the Euro 3 standard. A research question that this thesis will hope to address is the effectiveness of the Euro 6 technology at reducing the total NO_X emission factors of diesel passenger cars. This result will have implications for the predicted effectiveness of the London ULEZ.

Remote sensing measurements previously conducted are showing an increase in the total NO_X emitted as NO_2. This has gone from 15% to 30% between the Euro 0 and Euro 4 diesel classes and has been on a consistent rise sine Euro 0 for petrol powered vehicles (Carslaw & Rhys-Tyler, 2013; Carslaw, 2005). With the introduction of more and more stringent limit values on diesel powered vehicles manufacturers have had to get better and better at optimising their systems so as to pass the type approval process. Section 2.3.4 discusses the need for additional NO_2 in the exhaust to increase the rate of reaction in the catalyst systems however it is apparent that not all of this is being removed through later stages in the catalysis system. Furthermore the introduction of diesel particulate filters to reduce the PM_{10} and $PM_{2.5}$ emissions may also be causing an increase in f_{NO2} in diesel cars (Liu *et al.*, 2012). The fractional NO_2 emitted by petrol engines, whilst on the increase, still represents an increasing fraction of a decreasing total NO_X emission factor and should not currently be viewed as a cause for concern however it remains a factor that should be monitored in the future.

2.5 Additional Vehicle Emissions Measurements

As discussed in Section 2.4.5 the levels of pollution in urban environments are not decreasing. Additional methods are required to measure vehicle emissions so the cause of the decoupling of real world concentration measurements from stricter vehicle emission measurements can be understood. A number of different methods for measuring vehicle emissions are presented in this section along with their relative strengths and limitations.

2.5.1 On Road Tunnel Studies

Road Tunnel Studies (RTS's) are a widely used method for assessing real driving emissions. First described in the 60's (Chang *et al.*, 1981) and first implemented in 1978 (Pierson *et al.*, 1978), tunnel studies have been used extensively across the USA (Ban-Weiss *et al.*, 2008; McGaughey *et al.*, 2004; Pierson *et al.*, 1990), Europe (Brousse *et al.*, 2005; Laschober *et al.*, 2004) and world wide (Ma *et al.*, 2004). A wide range of species were measured including Nitric Oxide, Nitrogen Dioxide, Carbon Monoxide, total NO_X, Carbon Dioxide and Particulate Matter (Ropkins *et al.*, 2009).

In the tunnel study experiment the tunnel is treated as a dilution tube with samples drawn from both the air intakes and the air vents or sites near to these locations. The total pollution for a given species can then be measured based on the difference between the intake and vent values. The time resolution of these studies can be between 15 minutes to 24 hours (Ropkins *et al.*, 2009). Tunnels are ideally suited to this type of study as the closed system effectively isolates the air from external sources of pollution. The design of some tunnels lend themselves to the installation of monitoring equipment and often an air management system is also in place allowing an accurate assessment of air flow. Tunnels may often feature a transport control system that can give accurate vehicle flow and density data as well as potentially being able to give vehicle meta-data as well. If vehicle traffic meta-data is available it has been possible to apportion emissions to different subsections of the vehicle fleet. Tunnels also offer the logistics required to set up a wide range of the same laboratory equipment discussed in Section 2.4.2.

Tunnel studies have proven to be successful at measuring the total output of the fleet and regression techniques have been applied to attempt to assign emission factors to different sections of the fleets (He *et al.*, 2006; Stemmler *et al.*, 2005). Despite the success and widespread use of this method there are some significant shortfalls when attempting to translate the results into urban environments. The tunnels are not representative of the road network found in urban environments and hence the vehicle dynamics are not representative of urban driving. Attempts to assign pollution emission factors to subsections of the vehicle fleet lacks the measurement of individual vehicles present in other methodologies and therefore any assertions based on this data require further validation from other methodologies.

2.5.2 Portable Emission Measurement Systems

A Portable Emission Measurement System (PEMS) is a device that is mounted into the back of a vehicle and measures the vehicle emission output as it is driven either on road or over a laboratory controlled drive cycle. The vehicle emission is sampled through a probe introduced into the exhaust air flow where it either measures the pollutant or subsamples it for analysis in another onboard system. There are a number of commercial PEMS available such as such as Semtech-DS PEMS systems, Clean Air Technologies Internationals ULH-2100-DGTM (El-Shawarby *et al.*, 2005), Horibas OBS series of instruments (HORIBA, 2004, 2006; Nakamura *et al.*, 2002) and Temets Gasmet FTIR Emission Measurement System (Daham *et al.*, 2005b).

Traditionally PEMS based studies have focused on whole journey measurements as they remain the legislative standard however with the introduction and commercialisation of accurate real time monitoring systems more recent research has become focused on the interpretation and implications of real time vehicle emission measurements (Ropkins *et al.*, 2009). NO and NO_X are typically measured using chemo-luminescence (Weaver & Balam-Almanza, 2001) however the robustness of theses systems can be called into question because of the interference of water with the NO_X measurements. Non-dispersive Ultraviolet (NDUV) systems (Sensors, 2006), electrochemical cells (Vojtisek-Lom & Allsop, 2001) and solid phase sensors (Kihara *et al.*, 2000; Nakamura *et al.*, 2002) have all been used to measure NO_X in PEMS systems as well. Typically chemo-luminescence PEMS systems operate in concentrations of $0 - 5000ppm$ with a sensitivity of $1ppm$. Other systems operate in the same range however they are not as sensitive (Ropkins *et al.*, 2009). PEMS measurements have been evaluated in side by side laboratory comparisons and have been shown to be in agreement with conventional laboratory based monitoring systems (HORIBA, 2004; Nakamura *et al.*, 2002; North *et al.*, 2004; Prati *et al.*, 2015).

Research has been conducted using PEMS to understand the differences between real world and drive cycle emissions. Takada *et al.* (2002) shows that the difference between real world and the Japanese drive cycle NO_X emissions was double the drive cycle emissions for real driving emissions. Guenther *et al.* (1996) also showed that real driving emissions were higher than drive cycle emissions using a PEMS system. More recent work has been undertaken to better understand the causes of these increased emissions. Takada *et al.* (2005) investigated the emissions in real driving environments characterised by stop start events, overtaking and turning as well as engine mode over real journeys by a diesel freight vehicle in Japan using an Horiba ZrO_2 PEMS. Higher NO_X was observed at lower gears likely because of the EGR system operation being sub-optimal in these ranges. PEMS measurements have also been used to test the effectiveness of Euro 6 technology, a key research question in this paper, with O'Driscoll *et al.* (2016) testing 39 Euro 6 vehicles with a range of pollution removal systems and showing that on average the Euro 6 vehicles were still emitting 5.3 times the urban type approval value and 3.8 times the motorway type approval values.

PEMS studies have been used to validate vehicle emissions models based on engine maps. El-Shawarby *et al.* (2005) used a PEMS system to investigate the impact of engine mode on fuel consumption and emissions rates with the data also being used to validate the *VT-MICRO* model (Ahn *et al.*, 2002; Rakha *et al.*, 2004). An attempt was made to use PEMS based data to estimate emission rates in the Multi-Scale Motor Vehicle and Equipment Emission System (MOVES) developed by the US's Environmental Protection Agency. Although the study was described as a pilot it was shown that a modal binning strategy applied to the PEMS data was a useful solution for MOVES alongside Vehicle Specific Power (VSP) (Hart *et al.*, 2002). PEMS measurements have also been compared to the COPERT model and have shown that the average PEMS NO_X measurement is 1.6 times the COPERT estimate (O'Driscoll *et al.*, 2016).

PEMS studies have also been conducted to assess the best way to reduce vehicle emissions using traffic calming interventions (Frey *et al.*, 2001; Rouphail *et al.*, 2001). Daham *et al.* (2005a) attempted to quantify the impact of speed bumps compared to speed cameras using a Fourier Transform Infra Red (FTIR) PEMS study and found that NO_X increased by 195% with the introduction of speed bumps. Increases in CO_2, HC and CO were 90%, 148% and 117% respectively. The use of speed cameras was concluded to be significantly better at reducing vehicle emissions than speed bumps.

PEMS data when coupled with GPS data provides a very robust and recognisable method for analysing vehicle emissions. Driver behaviour, road environment and ambient conditions that could not be accounted for in laboratory based studies or tunnel based studies is can be measured and factored into the analysis. In vehicle measurements are generally thought of as the best tool for analysing the impact of various cases on vehicle emissions. The PEMS data sets do have significant limitations however. The number of vehicles measured using PEMS is limited by the cost of installing and the additional minor modifications that are required. It is therefore difficult to use the PEMS data collected to extrapolate over the entire fleet as the sample size is very small. The additional weight added by a PEMS system may introduce additional bias however the industry trend is going towards reducing the weight of the instrumentation. PEMS data cannot be considered representative of the fleet as a whole. Furthermore the problem of synchronising the PEMS data to the GPS or additional metadata is non-trivial. Often the offset is not a simple linear time offset but is a differential relationship to the process being measured. For example the vehicle specific power is not just dependant on engine speed but on a number of other factors such as driver behaviour, previous engine speed and inertia. These factors may not be linearly linked. In addition to this problem the measurement of the emission may not accurately represent the production of the emission. Physical interactions within the tailpipe such as diffusion, dilution and mixing of the exhaust gasses can all contribute to this misalignment of data (Ropkins *et al.*, 2009).

Whilst PEMS data remains a useful tool for understanding the emissions of an individual vehicle there is still room for improvement in the general field of vehicle emissions measurements. Cross-road or remote sensing studies, which will form Section 2.6, provides a useful tool that can be deployed in addition to PEMS studies and dynamometer studies to help understand exactly what is going on with vehicle emissions and attempt to provide strategies that can help to minimise the total emissions a fleet emits into the local environment.

2.5.3 Vehicle Simulation

The use of microsimulation packages such as AIMSUN or VISSIM which can output an individual trajectory or trajectories derived from vehicle tracking are becoming useful as vehicle emission modelling improves. These vehicle traces can be used as inputs to vehicle emission models that can provide emissions information similar to that achieved using PEMS methodologies.

The Passenger car and Heavy duty Emissions Model PHEM (Hausberger, 2003) takes a second by second vehicle trajectory and uses modal chassis and engine dynamometer tests to create emission maps that predict a vehicle's emissions based on the instantaneous speed and power demand as well as vehicle specific factors such as the size, weight, fuel type and euro class of the vehicle. Tate & Connors (2014) presented possibly the largest survey created in the world using the combination of vehicle emissions modelling (PHEM) and vehicle microsimulations (S-PARAMICS) in York, West Yorkshire. Vehicle traces were generated by the microsimulation model and fed to PHEM which was able to estimate the fuel consumption, NO, NO_2, CO HC, particle mass and particle number for tailpipe emissions of petrol and diesel fuelled vehicles of a range of Euro classes and operational vehicle types such as cars, vans, busses, coaches and heavy commercial vehicles. The data was further subset into time periods with the difference between AM, PM, free flow and night time driving analysed. The high temporal resolution of the microsimulation meant that the emissions model was able to map vehicles through intersections and junctions and show that at congested times the vehicle emissions were spread further along the street due to vehicles waiting in queues whereas at free flow times the vehicle emissions were focused around the junction controls themselves and concludes that pollution dispersion models would be better represented as a series of point sources at junctions rather than as line sources with uniform emission along the length of the link.

Initially these modelling methods have been shown to be unreliable, often giving errors in the order of ±50% (Joumard *et al.*, 1999) however since initial validation attempts the methodology has been improved. Wyatt *et al.* (2014) showed that the results from a PEMS system (Horiba OBS-1300) and the emissions predicted by GPS measurements inputted into PHEM for CO_2 show a strong positive correlation for the single vehicle tested in the study driven over repeated laps in Leeds, West Yorkshire, with the average PHEM result being 91% of the PEMS result. Kraschl-Hirschmann *et al.* (2011) showed that a combination of VISSIM and PHEM could be used as a first step to analysing whether or not a prospective traffic policy change has a noticeable effect on total vehicle emissions. The methodology has been extended to study heavy duty vehicles in port areas (Zamboni *et al.*, 2013) and Krajzewicz *et al.* (2015) was able to use PHEM to assess optimal vehicle speeds through traffic light controlled junctions.

PHEM has also been used as a tool for local emissions studies in a number of different cases (Tate, 2016, 2013a,b; Zallinger *et al.*, 2008) and has proven useful in assessing the total CO_2 emitted by a vehicle driving over a network of interest. The relative ease of data acquisition compared to PEMS data without the need to modify any vehicles means that this methodology is an exciting concept with the potential to be used in a number of different fields, some of which will be explored in this thesis, across the field of real driving emissions. The validation of the CO_2 measurements provided by PHEM is of particular interest in generating emissions factors using remote sensing data as the device it's self can only directly measure the ratio of a species to CO_2 and emission factor derivation (described fully in Section 3.3.2) requires assumptions about the CO_2 which affect the scope of use of the measurement.

2.6 Cross-Road Remote Sensing Methodology

2.6.1 Introduction

The following section will describe the history of remote sensing. It will look at the evolution of the device from initial stages through to the current prototype instrumentation.

Remote sensing principles can be applied over long (>100km), medium ($\approx 1km$) and short (<100m) path lengths. Short open path deployments have typically been set at ground level to capture exhaust plumes from passenger vehicles and vehicles with low mounted exhausts however some studies designed to capture elevated exhausts from Heavy Goods Vehicles and other vehicles with similarly positioned exhausts (Bishop *et al.*, 2001).

Data collected using the short path length Cross Road Remote Sensing methodology forms the majority of the data presented in this thesis. The Remote Sensing Device used by the Institute for Transport Studies (*henceforth referred to as RSD or RSD4600 when multiple systems are used*) is the first to be used extensively in the United Kingdom. It is a Remote Sensing Device model 4600 serial number

4612 and is manufactured by ESP systems. It is capable of measuring thousands of vehicle plumes per day and capture license plate information about the vehicles that it is measuring. The combination of license plate data acquired in post process and vehicle emissions measurements allows for the study of vehicle emissions as they relate to vehicle manufacturer, make and model, vehicle class, euro class and fuel type. This facility is not achievable with any other commercially available piece of equipment currently being used in the United Kingdom. Alongside this the FEAT system is a prototype instrument which can measure additional species as described Section 2.6.5.

Remote Sensing Device studies have been performed a number of times in the UK using either the FEAT prototype system that had been borrowed from the University of Denver or the RSD4600 used alongside the FEAT device (Carslaw & Rhys-Tyler, 2013; Carslaw *et al.*, 2011b,b, 2013). The Remote Sensing Device can also be used as part of a consultancy process for understanding fleet composition and contribution to pollution inventory for low emission zones (Tate, 2016, 2013a,b). Use of the Remote Sensing Device in these situations allows local governments to design more bespoke emissions strategies with relevance to their local fleet and effectively calibrate emissions models to accurately reflect their fleet. The overall aim of these strategies is to reduce pollution emissions and improve air quality in the urban environments.

The majority of experiments using open path cross road remote sensing devices have been performed using the devices developed by Stedman and Bishop, their co-workers or people using either their devices or commercialisation of their devices (Ropkins *et al.*, 2009). The data presented in this thesis has been conducted using the RSD4600 device, a commercialisation of the Stedman and Bishop System.

2.6.2 Initial Development for Carbon Monoxide

The first generation Remote Sensing Device designed to measure vehicle emissions measured carbon monoxide (Section 2.2.4 and carbon dioxide (Bishop *et al.*, 1989). Carbon monoxide emission of a vehicle depends on the air to fuel stoichometry and incomplete combustion of the fuel, where not enough oxygen is present to completely oxidise the Carbon to CO_2 will lead to Carbon Monoxide production (Bishop & Stedman, 1996).

To measure the Carbon Monoxide an infra-red source, located on one side of the road, sends a collimated beam into a gas filter radiometer equipped with 2 liquid nitrogen cooled indium antimonide photovoltaic detectors on the other side of the road. The signal beam is split into two and a $4.3\mu m$ bandpass filter is applied to isolate the CO_2 signal and a $4.6\mu m$ bandpass filter is applied to isolate the CO signal. The CO signal passes through a rotating gas filter wheel which modulates the signal through a CO and H_2 mixture channel and a N_2 channel creating a CO signal and a reference signal. The device is calibrated using controlled gas flow through calibration cells. The accuracy of the device was determined using experiments using electric vehicles emitting known controlled concentrations and volumes of pollution and performed at Bandimere Speedway with between 3 and 5 vehicle pass-throughs at 10, 20, 30 and 40 miles per hour and validated using an onboard emissions sensor and found to be accurate within measurement tolerances of $\pm 1\%$ (Bishop *et al.*, 1989). The Carbon Monoxide detector has also been proved able to measure vehicle emissions on road (Bishop & Stedman, 1990).

2.6.3 Second Generation RSD for Hydrocarbon Measurement

To improve on the Carbon Monoxide measurement capabilities of the Remote Sensing Device a Hydrocarbon module was later introduced (Guenther *et al.*, 1995). Unburnt hydrocarbons occur because the cold metal wall of the cylinder extinguishes the flame in a layer of the order 1mm thick known as the quench layer (Bishop & Stedman, 1996). This phenomenon is present in petrol vehicles using a spark ignition (LoRusso *et al.*, 1981) and is also present to a much more complicated level in Diesel engines where quenching can come from multiple sources including the fuel it's self (Yu *et al.*, 1980).

Significant modifications were made to the optical section of the previously described Remote Sensing Device to allow for hydrocarbons to be measured. Reflective as opposed to the previously used refractive optics are exploited. A twelve-sided polygonal mirror spinning at $12k$ revolutions per minute (rpm) is used as opposed to the beam splitters in the previous generation. The beam is split into four and sent to four different detectors corresponding to the 4 measurement channels each with their own interference filter for HC, CO, CO_2 and a reference channel. The conceptual change from the previous model is that each sensor now sees the whole signal for part of the time rather than part of the signal for the whole time. A wavelength of $3.3\mu m$ was chosen to measure hydrocarbons as excitation at this wavelength is due to stretching of hydrogen carbon bonds and dry air has no absorption at this wavelength. Propane has also been shown to absorb light of this frequency and alkanes are known to be strongest absorbers compared to aromatics. Measurements at this wavelength can be positively biased by the presence of gaseous water or condensed liquid water (steam) that is also emitted from the exhaust pipe (Guenther *et al.*, 1995). Selection of the frequency window is chosen to minimise the impact of this.

The hydrocarbon element has been tested extensively. Experiments using instrumented vehicles with variable air to fuel mixture were able to show that the HC element of the Remote Sensing Device was accurate to ±15% of the instrumented vehicle measurement (Ashbaugh *et al.*, 1992). Tests performed on road were successfully able to identify vehicles that were emitting a significantly high amount of HC, referred to as *high emitters*, on the road (Zhang *et al.*, 1993, 1996b).

2.6.4 Third Generation Addition of Nitric Oxide Measurements

The results presented in this thesis will focus on the emissions of Nitric Oxide (NO) and total NO_X from vehicles. The RSD4600 measures NO directly and NO_X can be estimated from this value using measurements of NO_2 provided by the prototype FEAT system (Section 4.3. This subsection will deal with the measurement of NO with further subsections detailing and critiquing the NO_2 estimation process.

An ultraviolet source was added to the Remote Sensing device to allow it to measure NO. A window centred on the NO absorption doublet at $226.5 \times 10^{-9} m$ (sometimes referred to as 227×10^{-9} in literature) to avoid the positive bias caused by the absorption due to the presence of water. The optical system remains the same as in the previous iteration of the device (Zhang *et al.*, 1996a). At the time of publishing it was understood that Nitric Oxide represented the majority of total NO_X present in exhaust gasses (Heywood, 1988). The 3000 iteration of the technology (RSD3000) has been used in numerous studies world wide (Chan & Ning, 2005; Chan *et al.*, 2004).

In later iterations of the remote sensing device the ultraviolet element of the Remote Sensing Device was upgraded to a high pressure xenon arc-lamp. The instrument has a spectral range of $16 \times 10^{-9} m$ and a resolution of $0.3 \times 10^{-9} m$. The instrument investigates the range $218 - 234 \times 10^{-9}$ meters and measures the same peak as the previous iteration of the device and has been used to show potential interference due to aromatic hydrocarbons present in the exhaust (Popp *et al.*, 1999a). The success of this iteration of Remote Sensing has been significant and they have been used in many long term studies as well as short term studies in the USA (Bishop & Stedman, 2008; Bishop *et al.*, 2010).

2.6.5 Remote Sensing of Nitrogen Dioxide

Given the concern about the amount of NO_2 in the plume (Carslaw & Beevers, 2004) and the potential for Ammonia slip from modern Selective Catalytic Reduction systems (Koebel *et al.*, 2000), the Remote Sensing Device have been updated to be able to measure these new species. The optical configuration remains the same as previously with the addition of another spectrometer to measure the greater range now required. The additional features are present in the FEAT research instrument used by the University of Denver however they are not present on the RSD4600 used by the University of Leeds. The additional features are fully described elsewhere (Burgard *et al.*, 2006) and summarised below.

Nitric Oxide, Ammonia and Sulphur Dioxide can all be measured across similar wavelengths and require improvements to the sensitivity of the UV detector so it can detect down to 200 nanometers ($10^{-9}m$). NH_3 has five absorption features between 200 and 220 nanometers and SO_2 has an array of features in the same spectral window.

The frequency chosen to measure NO_2 is peaked at 438×10^{-9} and is the location of the largest NO_2 absorption feature free of interference from other species. Modification of the initial UV spectrometer to accommodate this additional frequency required a simple adjustment of the grating position.

2.6.6 Remote Sensing of Particulate Matter

The first particulate matter measurements were made available with the RSD4000 instrument in 2002. Particulate matter is measured by observing the opacity of the emissions plume using an ultraviolet source in the RSD4600 and an infrared source in the FEAT system. A smoke number can be calculated based on the change in opacity. An ultraviolet source is more desirable for calculating the opacity of diesel exhaust emission as the wavelength of the light source is comparable to the cross section of the particles to be measured. The infrared source is less desirable as it will not interact with the smaller particles that the UV source would interact with.

The wavelength chosen for the ultraviolet smoke factor is $\approx 232nm$ and is chosen because the wavelength corresponds to the peak mass density of exhaust plume. A smoke number (*uvSmoke*) is calculated and translates to the number of smoke particles per unit of fuel. The smoke number is calculated in Equation 2.10.

$$uvSmoke = \frac{-100 \times ln(T_{UV})}{N_{CO2} + N_{CO} + N_{HC}} \tag{2.10}$$

This methodology has proved successful at identifying high smoke emitting diesel cars and has the potential to also identify highly emitting petrol vehicles as well.

2.6.7 Remote Sensing Device Usage

Historically many of the early experiments using remote sensing focused on testing for EPA conformity in the 1970's and 1980's in Arizona and then extended across to other cities in the USA using idling speeds and constant engine speeds to attempt to identify malfunctioning or especially dirty vehicles (Ropkins *et al.*, 2009). Studies were then to show that the difference between mobile sources and measurements taken with vehicles running at fixed speeds were significant (Ashbaugh & Lawson, 1991; Bishop & Stedman, 1996; Lawson *et al.*, 1990; Mazzoleni *et al.*, 2004; Walsh *et al.*, 1996; Zhang *et al.*, 1996b). These differences meant that the measurements of emissions of modern vehicles could not be well assessed using the remote sensing device and it was also shown that the RSD was not useful at identifying the super highly emitting or very dirty vehicles that it was primarily designed for. In contrast to this Stedman *et al.* (1993) was able to show that a 50% or greater reduction in CO was observable when drivers of very dirty vehicles were offered free maintenance of their vehicle compared to a 14% reduction in a control group comprised of high emitting vehicles where the owners were not informed.

Attempts have been made to use the RSD methodology to compare vehicles that emit highly in RSD measurements to their performance over the IM240 drive cycle. The IM240 drive cycle is a short standardised chassis dynamometer test based on the FTP75 drive cycle, lasting 240 seconds and covering 3.1 kilometres. The top speed is $91.2kmh^{-1}$ and the average speed is $47.3kmh^{-1}$. The IM240 drive cycle is used by the US EPA as part of it's inspection and maintenance (IM) program. Initially Stedman *et al.* (1994) showed that there was agreement between methodologies with those vehicles identified as high emitters producing extremely high levels of emission over the IM240 drive cycle. Further work using larger data sets confirms that there is correlation between those vehicles that emit highly in remote sensing studies and those that emit highly over the IM240 drive cycle (Pokharel *et al.*, 2000a,b).

The use of the remote sensing device to study the distribution of the fleet as a whole is one of the most important and interesting outcomes of the methodology. Use of the remote sensing device is similar to that of the road tunnel methodology except that the emission can be tied directly to an individual vehicle (Ropkins *et al.*, 2009) and using vehicle registration databases it is possible to assign emission characteristics to subsets of the vehicle fleet Tate (2013a), Tate (2013b) and Tate (2016) all use this methodology.

Many studies have been performed in America to analyse the distribution of the fleets. In Colorado (Ostop & Ryder, 1989; Stedman & Bishop, 1990), Chicago (Stedman & Bishop, 1990), Los Angeles (Lawson *et al.*, 1990; Stedman *et al.*, 1994) and Utah (Stedman *et al.*, 1993) this analysis has been performed. It has been determined that in these cases only a small percentage of the vehicle fleet is responsible for the majority of the emitted carbon monoxide. The results are typically 10% of the fleet for 50% of the emissions. This analysis was repeated for HC where less than 15% of the fleet was responsible for 50% of the HC and CO emissions (Stedman *et al.*, 1994). Zhang *et al.* (1995) extended the analysis of HC and CO worldwide finding results that generally agreed with the previous results with less than 15% and sometimes less than 10% of the vehicles contributing greater than 50% of the total emissions in cities including Bangkok, Edinburgh, Gothenburg, Hamburg, Kathmandu, Lisbon, Lyon, Thessaloniki and Zurich. Popp *et al.* (1999b) further extended this analysis to include NO measurements observing the same trend. Carslaw & Rhys-Tyler (2013) also arrived at similar conclusions for NO_2. Stedman & Bishop (1990) and Zhang *et al.* (1995) also showed that there is a statistically significant ($R^2 \approx 0.74$) correlation between CO and HC emissions suggesting that those vehicles with high CO are also high HC emitters as well. The correlation between CO and NO was not observed suggesting that the contributors of HC and CO were different from those producing NO.

High emissions may be associated with faulty equipment (Stedman & Bishop, 1990). Studies have been performed using time series style measurements over a range of years to try and assess the impact that vehicle age has on it's emissions. An increase in emission with age for CO emissions (Lawson *et al.*, 1990) and also for HC and NO_X

has been observed (Popp *et al.*, 1999b). Bishop *et al.* also showed a decrease in NO_X performance with an increase in vehicle age but not the decrease in magnitude of CO and HC performance observed elsewhere. Pokharel *et al.* (2001) showed that a large increase in vehicle emissions was observed after a 7 year period however Bishop (2007) showed that the vehicle emissions were not statistically related to the vehicle age. Similar results were observed when mileage was linked to emissions with no real observable effect observed (Borken-Kleefeld & Chen, 2015). This result may be explained by the introduction of the US Reformed Gasoline Program in the test area however. The efficacy of these studies is subject to some doubt as unavoidable changes in the vehicle fleet have been beyond the control of the study leaders. Factors such as vehicle make and model, fuel type and composition and the physical parameters of the vehicles themselves all vary from one year to another however broadly speaking these trends provide a realistic snapshot of the fleet's behaviour over time (Ropkins *et al.*, 2009).

Remote sensing devices have also been used to attempt to validate emissions models. Ekström *et al.* (2004) attempted to evaluate the COPERT III model (Ntziachristos *et al.*, 2000). RSD measurements were generally in agreement with COPERT III when measuring NO_X for all cases of petrol and diesel powered passenger cars but less successful for CO where COPERT III overestimated the total CO. For HC emissions COPERT III had a small tendency to overestimate the total emissions. The COPERT III model noticeably underestimated the emissions by diesel heavy duty vehicles however and the reduction in NO_X from Euro 2 and Euro 3 was significantly less than predicted by COPERT III. Carslaw *et al.* (2011a) compared RSD observations to predictions made using the Handbook for Emissions Factors (HBEFA, 2010)) and showed that these emission factors were an underestimate for diesel powered vehicles.

The remote sensing in situ measurements are useful at assessing a local fleet's emission performance for a number of different pollutants and shown to have a range of different uses in the context of other studies.

2.7 Summary

The literature review presented in this chapter has examined the various chemicals that are considered to constitute air pollution in urban environments. Evidence is presented to show that there is significant negative health impacts related to the exposure to these chemicals. The list presented is far from complete, focusing on those that can be measured using the remote sensing device and secondary pollutants that are impacted by these primary pollutants. The review has also briefly assessed the impact of industrial sources not measured by the remote sensing device for context.

The literature review then assessed the impact of vehicles on air pollution and the steps to remove these harmful chemicals from the exhaust gas emissions. The chemistry underpinning the catalysts used in both diesel powered and petrol powered vehicles is presented. Three way catalysts are described and the chemical reactions and optimal engine and environmental conditions of operation are discussed. Lean burn diesel engine exhaust emissions removal systems are also discussed. The differences and associated challenges between lean burn and stoichiometric engines especially relating to the removal of NO_X are also discussed with the solutions currently being deployed in vehicles evaluated.

The legislative steps taken by governments at reducing ambient air pollution are presented. The use of drive cycle based type approval for new vehicles is discussed. The different drive cycles available to legislators in the EU and in the USA are presented alongside other drive cycles that have been developed for research and testing purposes. The WLTP, a drive cycle developed for future Euro 6b vehicles is also presented. The relevance of these drive cycles to real world driving is assessed using RSD measured data for speed and acceleration and the FTP-75 is shown to be the most representative of real driving conditions measured by the RSD. The Euro class legislation that applies to all cars sold in the UK is summarised and the effectiveness of these steps is assessed. The lack of previous success in predicting the effectiveness of introducing new technology via legislation such as the Euro standards or direct interventions such as the London Low Emission Zone are identified as gaps in research that could be filled at least in part by remote sensing data. Chapter 5 presents a framework for a remote sensing based assessment to be undertaken and Chapter 6 applies this methodology to the new Euro 6 vehicles that have been measured by the remote sensing device. Chapter 7 applies some of the lessons learned in previous chapters to examine other areas of the fleet in a more explorative way.

The various methodologies that are used to assess vehicle emissions are presented and assessed prior to the description of the remote sensing device. A selection of studies for each different methodology are presented with the various strengths and weaknesses being compared and contrasted. The remote sensing device is then described in depth. A historical development of the methodology is presented beginning with the first instrumentation developed in 1989 and finishing with the current state of the art technology. The instrument used throughout this thesis is shown in context as not being the current prototype state of the art equipment and the lack of NO_2 measurements required for total NO_X assessment are presented, forming the research question tackled in Chapter 4. The full data collection and methodology is presented in Chapter 3 alongside all of the meta-data associated with site selection.

The lack of a reliable modelling methodology for predicting the emissions behaviour of whole fleets using data collected in real driving environments is identified. Modelling of the distribution of vehicle emissions measured by remote sensing devices was attempted in one paper however not developed further. Chapter 5 attempts to further this research and develop it to the level where it combined with the work presented in Chapter 4 can be used as a predictive tool for NO_X emission in Chapter 6 and to test new policy selection impact on NO_X fleet emissions in Section 7.2.

Chapter 3

Remote Sensing Device Methodology and Data Collection

3.1 Introduction

The purpose of this chapter is to describe the remote sensing device and the data collection process. It details the theory of the physical processes which underpin the measurement, the limitations that this imposes and the process of taking a measurement with the remote sensing device. It discusses how the instrument is set up for the range of different configurations that were used throughout the data collection phase of this thesis and the additional data collection processes that were not automated through the RSD system. These additional processes were developed to improve the capture rate of certain types of vehicles not suited to automated data collection such as HGVs that still contribute NO_X to the emissions inventory.

The vehicle emissions data collected and presented in this thesis was primarily collected using a commercial cross-road remote sensing device referred to as an RSD4600 (Section 2.6). Additional data was collected using the prototype instrument whilst on loan from the University of Denver referred to as the FEAT system (Section 2.6.5). The following chapter builds on the background of the remote sensing device described in Chapter 2 by describing the physical processes that underpin the measurement and reviews the advantages and disadvantages of the remote sensing methodology. The chapter then reviews the operating procedure including site setup, calibration and auditing of the instrument as well as providing details of how each individual vehicle is measured. This chapter also describes each of the on-road locations where measurements used in this thesis were taken and finishes by summarising the data that was collected.

3.2 Theory

3.2.1 Interaction of Light and Matter

The remote sensing device takes advantage of the interaction of light and matter. The measurement of molecules in vehicle exhaust plumes requires an understanding how light from the remote sensing device interacts with matter in the exhaust plume. A photon of light has a defined and discrete energy E related to it's frequency ν (or it's wavelength $\lambda = c/\nu$ where c is the speed of light, $\approx 3 \times 10^8 ms^{-1}$). The relationship is $E = h\nu$ where $h = 6.626 \times 10^{-34} m^2 kg s^{-1}$ is Plank's constant (Planck, 1900). The molecules exist in their lowest (or ground) energy state wherever possible but can be excited into a higher energy level if they interact with a photon with the energy corresponding to the difference between energies of the energy levels of the two states. A photon with either too high or too low energy will not excite the electron into the next level so fine tuning of the light beam's energy can be used to probe matter and understand what it is made of. Historically this process is most successfully demonstrated in the photoelectric effect causing the ejection of electrons bound in metal sheets to form free electrons (Einstein, 1905). The same principles can be used to remotely determine the content of any gas cloud back-lit by a source of light. This concept has been used to understand the structure of the universe using Lyman Alpha transition for neutral hydrogen ($n = 1 \rightarrow n = 2$) (McDonald *et al.*, 2006) as well as many other physical applications and the same principles can be applied to gas phase molecules present in vehicle exhaust plumes allowing their constituent parts to be measured.

With knowledge of the energy level structure of the target molecule and the availability of technology to create photons of the correct energy it is possible to exploit electron capture to measure a column density σ defined in Equation 3.1 along a path l. Remote sensing devices are able to exploit this physical process by using a combination of tuned lasers and bandpass filters to create beams of photons with an energy corresponding to a known energy level of the target molecule. By observing the attenuation of the photon count from the source they are able to measure the photon capture by molecules present in the exhaust plume and understand the composition of it relative to other molecules. Typically the measurement of a pollutant such as NO is expressed in terms of a ratio between it and CO_2.

$$\sigma = \int \rho dl \tag{3.1}$$

The column density is derived using the Beer-Lambert Law that relates emitted and measured intensity changes as the light passes through a medium. The Beer-Lambert Law is defined in Equation 3.2 where I is the intensity of the light beam and τ is the optical depth of the medium the light passes through.

$$I = I_0 e^{-\tau} \tag{3.2}$$

To measure the molecular content of the exhaust plume the remote sensing device works in the most simple terms by shining a beam of light through a plume of gas and counting the number of photons that are absorbed by measuring the decrease in intensity at specific frequencies. This is performed for multiple species and with multiple frequencies of light. The relative abundances of each species can then be calculated. The equipment and it's evolution used is described in Section 2.6.

3.2.2 Advantages of Using A Remote Sensing Device

As described in Section 2.5 numerous methodologies have been used to quantify vehicle emissions each with their own advantages and limitations. Remote sensing device studies as performed by ITS have a number of significant advantages that cannot be achieved through other methodologies. As with any vehicle emission measurement methodology compromises have had to be made. This has also lead to some limitations as well as some advantages. The advantages and limitations of the RSD are discussed in the next two subsections.

The most important advantage of using remote sensing relates to the sample size of the population surveyed. Remote sensing allows the emissions of all vehicles driven by their own drivers on real roads to be measured directly. Laboratory and PEMS studies are expensive, labour intensive and limited in the vehicles they can measure (Sections 2.4.2 and 2.5.2) . For example Marotta *et al.* (2015) compared drive cycle emissions of 21 vehicles and Favre *et al.* (2013) were able to measure six vehicles over the NEDC and the WLTP on chassis dynamometer tests, Andersson *et al.* (2014) was able to test two vehicles using laboratory tests and PEMS measurements, Kousoulidou *et al.* (2010) was also able to measure six vehicles in a study using a PEMS system and Weiss *et al.* (2012) analysed PEMS measurements from 5 vehicles, one of which was a novel Euro 6 diesel vehicle. In comparison a remote sensing device can measure greater than 2×10^4 vehicles over a week long survey. Hiring a basic chassis dynamometer is expensive. AET Motorsport price their dynamometer at £65 per run for a two-wheel drive vehicle or £80 an hour without a technician. Additional labour and equipment required for the vehicle emissions measurement section of the test, which is strictly controlled, will only add additional cost.

Due to the expense of testing in a laboratory the number of vehicles tested is often low and funding is typically only available to test the newest technologies. Testing of old fleet vehicles over regular time intervals to assess any impact of ageing is rarely performed. The cost required to test the emissions of vehicles means that further potentially influential factors are not accounted for in laboratory tests. Factors such as cold starts, vehicle payload, tyre type and pressure, 4 wheel drive systems, enthusiastic amateur aftermarket modifications and engine remapping are rarely tested in the laboratory are accounted for. Given the small number of vehicles sampled in laboratories if there is a problem with the vehicle to be tested it might be seen as a particularly high emitter in class. Factors such as faulty vehicles and failed catalysts (Section 2.3.2) would not often be observed in laboratory tests of golden vehicles (Section 2.4.2) but would be encountered during a remote sensing survey. If vehicles supplied to the laboratory do include faults the results would be biased towards higher emissions. Vehicles that are emitting more than expected can be compared to a larger number of fleet vehicles to see if this a standard result or a one-off issue with the lab tested vehicle. PEMS studies require minor modification to the vehicle to run and this significantly limits the number and availability of vehicles on which such studies can be performed (Section 2.5.2) and this would still not be able to account for all of the influencing factors described. The RSD can be rapidly deployed and is able to assess the impact of new technology in ways that the PEMS and laboratory tests are not.

The meta-data collected through the use of captured license plate data cross referenced with vehicle registration databases allows analysis of vehicle characteristics such as make, model, fuel type, euro class and year of registration as well as the vehicle CO_2 rating that is used to calculate the emission factor in Section 3.3.2. Along with vehicle meta-data the speed and acceleration is also measured. This data allows for the broad strokes provided for by the volume of vehicles measured to be refined further. Not only does this type of project provide a more realistic snapshot description of the whole fleet it can also be used to assess exactly where the majority of the pollution is generated. It will be shown throughout this thesis that the remote sensing device can also identify vehicles that are behaving atypically from vehicles of a similar fuel type, vehicle class and euro standard. The ability to precisely record a snapshot of the fleet at a given location is something that no other methodology can achieve without making significant estimates and extrapolations from smaller data sets.

3.2.3 Limitations of Remote Sensing Device Study

As with all methodologies certain compromises have to be made. There are a number of limitations with the use of Remote Sensing Devices for measuring vehicle emissions. Site selection, instrument precision and the geometry of the tailpipe position on the measured vehicle can all limit the data captured by the instrument. These issues will be discussed and the steps taken to mitigate them described in this subsection.

The site selection needs to satisfy a number of criteria for remote sensing to be successful. Roads need to be single carriage as the RSD sees the total emission across the whole path length of the light beam and therefore is unable to distinguish between different vehicles if they are running side by side such as would be expected on a dual carriageway. This limits the number of sites that can be selected for monitoring however the number of locations that are satisfactory remains high in most urban centres. A range of sites need to be observed to ensure that the vehicle dynamics are as best a representation of the whole network as possible. This prevents any location specific features from biasing the overall results. It is desirable but not necessary for the site to have traffic calming structures such as central dividers or pedestrian refuges to facilitate the safe positioning of the instruments and the support vehicle for ease of deployment and to reduce the risk of damage to the instrument caused by a vehicle colliding with the instrument. Safety cones are extensively distributed around the instrument to mitigate this factor but this concern also limits the sites available for selection.

The ideal site is located on a slight positive gradient, typically between 0° and 3°, after a junction or both. The Remote Sensing Device requires an exhaust plume to take a measurement so sites where the vehicles are off throttle, cruising or decelerating result in a low percentage of valid measurements. The exhaust plume is either not present or too insignificant to be observed. The presence of road gradient or a junction layout means it is more likely that the throttle will be applied at the time of measurement and more emissions will be present in the plume leading to a higher capture rate. The requirement for engine load to be positive has the potential to introduce a positive bias to the results however the site selection of the studies conducted in this thesis has been performed in such a way that they are representative of the network as a whole as best as possible. This is examined in Section 3.3 and for example the site selection in Cambridge mostly comprised of flat roads as would be found in the city whereas the study performed in Sheffield had sites with more variation in gradient, more representative of the non-homogeneous distribution of gradients over the whole network.

The Remote Sensing Device is portable but is restricted to taking a snapshot of the vehicles that pass through the site rather than measuring the vehicle as it travels across the entire network. By selecting multiple sites across a network for RSD study this limitation is mitigated. As the number of measurements increases the range of operating conditions increases. By taking hundreds to thousands of measurements of the same subsection of the fleet (for example Euro 5 diesel passenger cars) a range of operating conditions representative of the network as a whole can be observed. Compared to Portable Emissions Measurement Systems (PEMS) however the RSD cannot cover the same amount of the network and cannot directly account for journey length and cold starts. For a more complete understanding of the whole network additional vehicle tracking measurements must be used.

The precision of the NO_X measurements taken using a Remote Sensing Device is significantly lower than laboratory based tests. A variance of 18% was observed in controlled repeated tests of the RSD4600 used throughout this thesis (Section 4.2.4) compared to the accuracy of (for example) the Semtech Ecostar which has an accuracy equal to the larger of ±2% of the measurement or ±0.3% of the total scale. The repeatability of the measurements is also lower than in a laboratory as vehicles other than busses do not typically drive repeatedly around urban environments. The variances of the instrument measurements and the vehicle emissions are compared in Section 4.2.5. The measurement error in the individual measurement means that the RSD is unsuitable for statements about individual vehicles with one or a small number of valid measurements in the same way that traditional high precision measurements would be. The RSD can however measure considerably more vehicles in a day than is possible in laboratory studies of individual vehicles and is ideally suited to understanding how the population of the fleet behaves as a group.

There are a number of geometrical design issues with vehicles that limit the ability of the Remote Sensing Device to measure them. Heavy Goods Vehicles are often high emitters and worth studying however a Remote Sensing Device cannot always generate valid measurements by it's self. Vehicle with elevated exhausts cannot be measured by a remote sensor unless it is elevated on scaffolding (Bishop *et al.*, 2009). Elevating the RSD means that it cannot measure vehicles with exhaust pipes positioned at kerb level. In studies designed to look primarily at passenger cars the addition of scaffolding is detrimental to the success of the project and was not used throughout this thesis. For articulated HGV's that do have an exhaust located at ground level the exhaust pipe is located behind the cabin but before the trailer. As the RSD camera takes a picture as it sees the peak exhaust plume concentration the result is often a picture of the side of a truck. If the geometry and license plate position is favourable the triggered image will include a license plate attached to the cabin however the license plate is not always located in the field of view of the camera. Additional observation strategies need to be undertaken to ensure that as many HGVs are captured. These strategies can range from the addition of a GoPro^{TM} camera constantly recording or a manually operated camera that captures an image of every vehicle that might be missed by the automatic system (Section 3.3.4). In both of these cases the additional license plate information is used alongside the triggered RSD camera which contains information about the type, colour and any other identifying marks to improve the completeness of the observed vehicle list. These additional methodologies were developed as part of this thesis.

From an instrumentation point of view the ideal time for taking measurements is during the summer season on weekdays during term time between the times of 0800 and 1800 as this window covers both the AM and PM traffic peaks that are of interest to policy makers and provide a large number of observations. Weekdays on term time represent the most important times where road usage is at it's peak and is the most representative of the sites studied. It is necessary to power up the instrument at or before 0715 to allow for the $45min$ warmup period to occur prior to commencement of measurements. Potential hazards of working on or near to a road mean that summer is the only reasonable time to take measurements as it is the only time when there is sufficient daylight available. Performing initial setup, alignment, calibration and audit (see Section 3.3.3) stages which require working in and repeatedly crossing the road puts the operators at significant risk of injury and summer months have the widest window in which to operate safely. Summer operating also minimises the impact of inclement weather that would impair the ability of the instrument to function. Anything more than a brief light shower requires the instrument to be covered up as it is not waterproof. Significant precipitation requires the instrument to be returned to the support van until such time as it can be returned to the road without risk of damage. As with all electronic instruments that have not been weatherproofed the introduction of water into the unit can cause significant damage. In addition to the direct damage caused by water the close proximity of the instrument to the road means that it is vulnerable to being exposed to the dirt. When mixed with rain the dirt-water mixture can easily coat the high precision mirrors and lenses that align the optical path of the sensing laser. When these are left to evaporate the remaining dirt can significantly reduce the efficiency of the optical path to the point where the device becomes unusable and requires servicing. It is not enough to simply polish the mirrors by hand as this would cause additional damage to the equipment. ESP Technologies have developed an environmental enclosure for the precision mirrors made from sapphire glass which prevents such damage however throughout the course of the data collection this upgrade was not available on the RSD4600 used. High wind speed can rapidly mix or disperse the exhaust plume resulting in a lower proportion of valid measurements as the pollution concentration in the plume has been diluted. It can also transport and re-suspend dust from the road. It remains unclear what the influence of wind is on the measurements however taking measurements on days with good weather serves to mitigate this effect prior to deployment. No measurements presented in this thesis were taken under extreme wind conditions.

The summers in the United Kingdom are not always as hot as those in other locations however the impact of environmental factors on the vehicles cannot be studied as well as would be liked. In colder conditions there are likely to be more vehicles operating under cold start conditions and not being able to measure in the winter months reduces the potential for observing these vehicles. Observations taken were not during peak times and therefore even less likely to capture freshly started vehicles. In addition the safety requirement for setting up when it is light means that the AM peak vehicles are missed and further limiting the potential to observe cold starts. The number of and contribution by cold start vehicles to the total emitted pollution is not known. Observing these vehicles in situ would be a first step to understanding these two factors however the RSD data collected throughout this thesis struggles to directly test these hypotheses. Attempts have been made in Section 5.4.2 to quantify the cold start fraction statistically however the results remain inconclusive.

3.3 Data Collection

Data was collected using the Remote Sensing Devices in a number of different configurations. Other ancillary devices were used to collect additional data to be combined with the Remote Sensing measurements. This section explains the methodologies used throughout the rest of the thesis and discusses the various strengths and weaknesses of each. Detailed descriptions of the data used for each of the results described here will be presented as the experiments themselves are completed in future sections. This section will also provide a summary of the five studies that were completed over the course of data collection for the thesis and introduce the new concepts developed during each one. Site descriptions, maps and photographs are also presented and annotated to show the details described in the schematic diagrams and descriptions applied in the real world.

3.3.1 The Remote Sensing Device Measurement

Raw Measurements

The Remote Sensing Device measures the area density ($N_\sigma = \int \rho ds$) of the exhaust plume. It is therefore independent of the geometry of the plume but is unable to give any information related to the number density ($N = \int \rho dV$) of molecules in the plume. Even though the remote sensing device cannot explicitly measure the total number of particles of a given pollutant it can be used to measure the relative abundances of different pollutants. The raw measurement from the Remote Sensing Device used in this thesis is a dimensionless ratio between Carbon Dioxide (CO_2) the test pollutant, either nitric oxide (NO) carbon monoxide (CO) or particulate matter (PM). The ratio is useful in it's own right as it is related to the efficiency of the pollution removal and reduction system however with knowledge of the CO_2 emissions of the vehicle an estimate of the absolute emission factor of NO can be estimated for a vehicle. A number of different methods are used to estimate the CO_2 emission factors for different vehicles and purposes. The different methods for calculating the CO_2 emission are discussed in Section 3.3.2.

The laser beam runs constantly until a vehicle passes through the instrument. A measurement is initiate by a block of the beam caused by a vehicle body. After the beam block $0.5s$ of data are collected at a sampling rate of $50Hz$. If the measurement is interrupted by, for example, another vehicle passing through the beam, then the measurement is invalid and discarded. If insufficient CO_2 ($< 5\%$) to make a measurement is observed in the plume then the measurement is also discarded as invalid. A valid speed and acceleration measurement in the range $5 - 60 kmh^{-1}$ is also required. To calculate the background levels of pollutants an average of the 5 measurements prior to the vehicle blocking the beam are taken and subtracted from the tailpipe measurements.

Calculating Total NO_X

The air quality legislation and vehicle emissions limit values are expressed in terms of total NO_X and an assessment of this emission factor is more useful than the NO emission factor alone (Section 2.4). Total NO_X emission is defined as the sum of the oxides of nitrogen present in the exhaust plume. This is generalised to $NO + NO_2$ since Nitrous Oxide (N_2O) is not a major factor (Becker *et al.*, 1999). The RSD4600 does not have the capability to measure NO_2 and therefore the fraction present in the exhaust plume (f_{NO_2}) needs to be estimated from other sources. A number of papers have done work to estimate the f_{NO_2} (Grice *et al.*, 2009) and to measure it directly (Carslaw & Rhys-Tyler, 2013). Regardless of the method of acquiring the f_{NO_2} measurement for vehicles the calculation of the total $NO_X : CO_2$ remains the same and is defined in Equation 3.3.

$$NO_X = NO \times \frac{1}{(1 - f_{NO_2})} \tag{3.3}$$

In previous work the NO to NO_X estimation has been performed using NO_2 values sourced from the literature derived using the *Netcen* roadside model based on the interactions of NO, NO_2 and O_3 (Abbott & Stedman, 2005; Grice *et al.*, 2009), a method commonly applied in such calculations, however this methodology requires a lot of assumptions and does not directly measure tailpipe NO_2 emissions.

The opportunity to validate the NO measurements with a similar instrument capable of measuring NO_2 directly would allow both NO_X estimates made using Grice *et al.* (2009) f_{NO2} estimates to be compared to total NO_X measurements and the measured values from the Carslaw paper to be combined with measurements taken using the RSD4600 for methodology comparison. The experiments assessing the viability of this method will constitute a part of this thesis and are described in Section 4.3.

3.3.2 Calculating Emission Factors

Derivation of Emission Factor

The emission factor is a useful metric to observe as it is directly related to all the legislations governing air quality. The Remote Sensing Device does not measure the emission factor, the concentration of or number of NO molecules directly but instead it measures the ratio of NO to CO_2 in the vehicle exhaust plume (Section 3.3.1). Given the $NO : CO_2$ ratio it is possible to derive an estimate of a vehicles NO emission factor each individual vehicle pass by. Equation 3.4 shows the process for calculating the NO emission factor and Equation 3.5 shows a generalised case for total NO_X emission factors.

$$NO_{EF} = \frac{NO_{RSD}}{CO_{2-RSD}} \times 0.68 \times CO_{2-EF} \times 10^{-4} \tag{3.4}$$

$$NO_{X-EF} = ([(1-f_{NO2})\times 0.68]+[f_{NO2}\times 1.045])\times \frac{NO_{RSD}}{1-f_{NO2}} \times CO_{2-EF} \times 10^{-4} \tag{3.5}$$

A factor of 10^{-4} is introduced to ensure that the units remain as gkm^{-1} (Carslaw & Rhys-Tyler, 2013) and a factor of 0.68 is introduced to account for the difference in mass between an NO molecule ($30.01gmol^{-1}$ and a CO_2 ($44.01gmol^{-1}$). The mass of NO_2 is $46.0055gmol^{-1}$ and the ratio of $CO_2 : NO_2$ mass is 1.045 and this factor is included in Equation 3.5 to ensure that the units are correct. The CO_2 emission factor will usually have units of gkm^{-1} however the user is free to create their own CO_2 emission factors that best suit their purpose. Units for the emission factor in terms of grams per litre of fuel burnt are often used in other sources. Grams per kilometre is the unit used in emissions legislation so for this thesis unless otherwise stated units of gkm^{-1} will be used for emission factors. Leaving the measurement as a ratio of the column densities of the two gasses is a useful result in certain cases as well. The ratio allows for a comparison of the efficiency of the pollution control systems and for some results presented in this thesis this method is preferred. Wherever this is used it is indicated that the ratio is the result.

Carbon Dioxide Estimations

A commonly used methodology for calculating emission factors this thesis is to base them on their manufacturers fuel efficiency rating which is obtained from the license plate meta-data. This methodology provides a real range of CO_2 emission factors that relevant to the wide range of vehicles observed. There have been a number of studies done using real world travel data rather than controlled lab tests, which can give unrealistic values for on road performance, on both passenger vehicles Ligterink & Eijk (2014) and light goods vehicles Kadijk *et al.* (2015). The results of these studies show that the manufacturer CO_2 values need to be uplifted by a factor greater than 1 to reflect real world CO_2 emissions. The CO_2 value used in the emission factor calculation (Equation 3.4) is shown in Equation 3.6 where ϕ_{CO_2} is the uplift factor provided by the appropriate literature for each individual vehicle class and expressed in decimal form such as $1.1 = 110\%$.

$$CO_{2_{EF}} = CO_2 \times \phi_{CO_2} \tag{3.6}$$

The uplift factor must be generalised in this case as it is impossible to measure the CO_2 emissions factors for each individual vehicle in the fleet. Whilst a limitation for comparing individual vehicles it remains a useful tool for comparing populations of vehicles and subsets of those populations where the data is available. Ligterink & Eijk (2014) provides a large scale ($n = 25 \times 10^3$ vehicles per year) study covering a wide range of vehicles and operating conditions. A number of different studies from a range of European countries (Netherlands, Germany, UK and Switzerland) were combined to achieve an overall European value for the uplift factor required as well as results more relevant to individual countries . Wherever possible the *Honest John* survey which contains data collected for UK vehicles ($n = 6500$ per year) and presented as part of Tietge *et al.* (2015) was used as it is the most relevant to UK roads however it was not always possible to use this data. In this case the choice of study used was based on the best availability of the data. In cases where no data was available, for example vehicles registered in 2015, the factor was set as the closest reported value, 2014 vehicles in the previous case. This is a significant limitation but represents the best educated guess at the time of writing. Extensions of the studies mentioned will allow future work to validate the emission factors that have been estimated using extrapolated CO_2 data. Given the general upward trend in the correction factors over the range and age of vehicles observed the assumption that the additional unknown fuel usage for future years would be at least as high as the last year is acceptable. The introduction of new technologies that may, and as yet unchecked, reduce the rate of increase of fuel consumption mean that it is unreasonable to attempt to extrapolate a trend. The fuel consumption change year to year is not dependent on previous values so extrapolating from that point forward using a model would not result in a better estimate so to avoid overestimating the result the last known value is the best compromise however future confirmation of this is required.

For light commercial vehicles (Section 7.3) Kadijk *et al.* (2015) used on road measurements to measure the emission factors of ten light commercial vehicles with varying mass. The results from this study are used for LCV emission factors. Euro 6 passenger vehicles are assigned an uplift value based on central estimates from Mind The Gap (Archer, 2015) which uses data collected from $n = 6 \times 10^5$ vehicles. This estimate does not fully take into account the change in technology introduced with Euro 6 so is potentially a source of some error in the calculation. Heijne *et al.* (2016) have direct measurements of Euro 6 CO_2 emission in Dutch urban conditions ($219 - 244gkm^{-1}$) however these are not transferrable to the methodology used here directly. Until such time as the Mind the Gap study present this result the central estimate currently used is the best estimate available however adjusting the results of the Euro 6 emission factors when better data is available may prove useful.

The emissions model PHEM (described fully in Section 2.5.3) estimates of CO_2 emissions can also be used to calculate a NO emission factor. These estimates have been shown to match PEMS data (Wyatt *et al.*, 2014). This methodology is useful when calculating emission factors for individual vehicles however it does not take into account the variance between manufacturers' CO_2 measurements when applied to the whole fleet. There are cases in Chapter 7 where PHEM estimates of CO_2 are used. PHEM allows for the calculation of CO_2 emissions for specific types of vehicles or individual vehicles if the data is available. When used as a source of CO_2 for calculation of NO_X emission factors an appropriate standard vehicle, based on Euro class and fuel consumption, is chosen as the modelled vehicle. These are dealt with on a case by case basis for different sections where applicable and the chosen parameters are stated in each section as they are introduced.

Once the CO_2 factor has been calculated it can be combined with the NO or NO_X emission measurements through Equation 3.4 to create an emission factor. The emission factor provides a real estimate of the amount of NO or total NO_X a given vehicle is emitting. Alongside the efficiency measurements provided by the raw ratio the emission factor derived from Remote Sensing Device measurements provides great insight into the ability to process NO_X of the vehicles as a fleet.

3.3.3 Experimental Design

A number of different configurations of the RSD are used throughout the course of the data collection. This subsection provides a detailed description of the differences between the three main types and additional details. Throughout the rest of the thesis the site description will reference one of the descriptions provided here. In all cases after the Remote Sensing Device is set up the equipment is turned on and left for a minimum of 45 minutes to reach thermal equilibrium. The instrument is then calibrated using internal gas cells of known concentration. An initial audit is performed using a control gas with known relative molecular abundances similar to those found in a petrol engine exhaust plume to ensure the accuracy of the calibration. The audit procedure is designed to mimmic a vehicle passing through the equipment whilst introducing a puff of control gas into the laser beam path which is measured by the equipment in the same way it would measure a vehicle. Once these steps have been completed the Remote Sensing Device is ready to take measurements.

Standard On-Road Deployment

The standard configuration used in all the on-road studies consists of the equipment described in Section 3.3.3 with the addition of a camera and speed and acceleration modules shown schematically in 3.1 and annotated in Figure 3.7. The speed and acceleration modules are situated approximately 2 meters up stream of the Remote Sensing Device. The support van is parked at a close-by location where the instrument communication and power cables can be taken from the

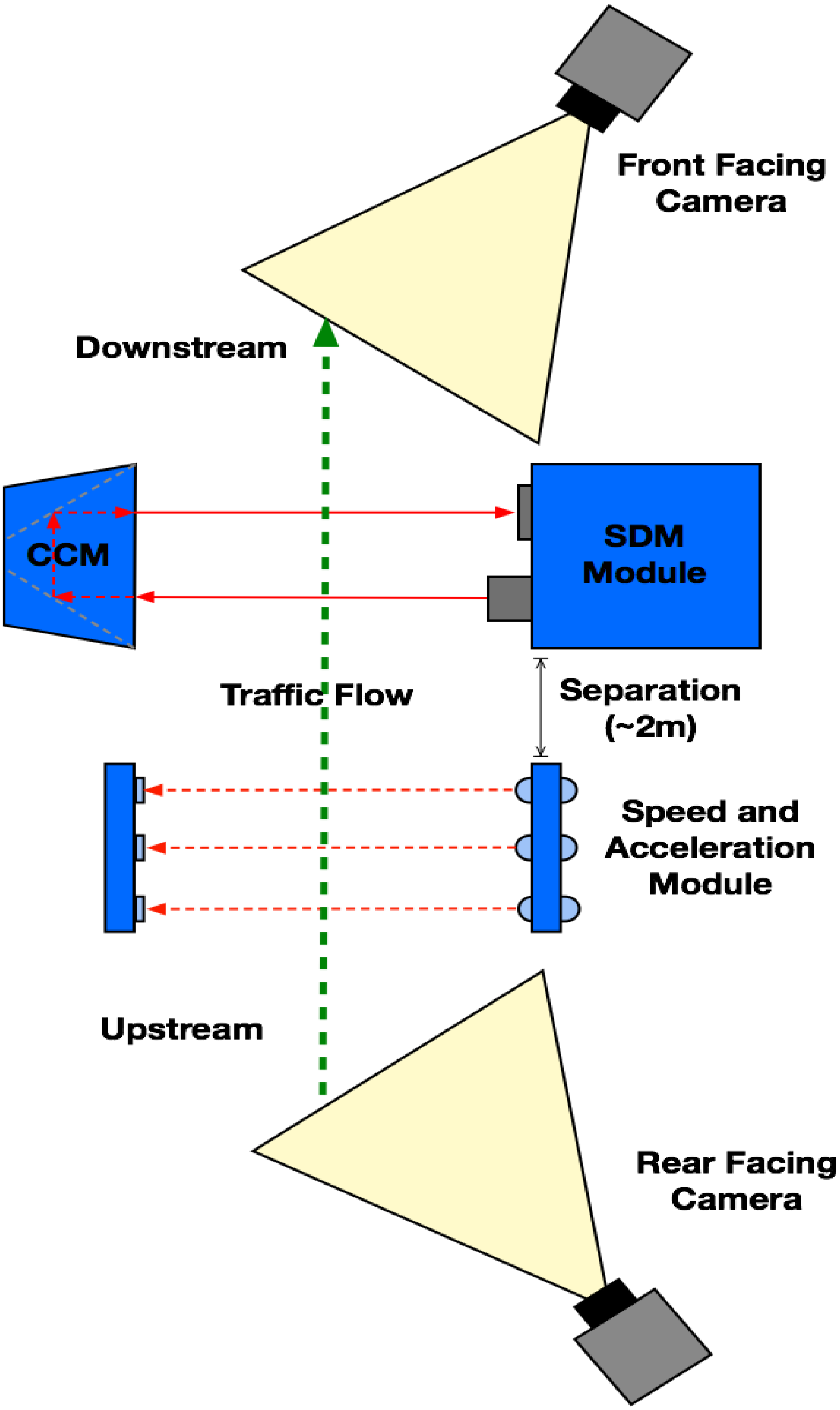

Figure 3.1: Schematic of the on-road experimental setup

SDM to the control computer and battery bank in the support van. Where necessary anti-trip devices are installed to protect the user and pedestrians from injuring themselves or damaging the equipment inadvertently.

The camera can be positioned in a front license plate facing or rear license plate facing configuration, both of which are shown in Figure 3.1. The front facing configuration can be seen annotated in Figure 3.6 and the rear facing configuration can be seen annotated in Figure 3.12. Selection of the orientation of the camera is decided firstly by the nature of the site. If there is only one way to locate the camera safely and without blocking off road user sight lines then that configuration must be used. If both options are available the vehicles that use the site will be the determining factor. Bus license plates cannot be seen from the front facing camera and HGV's license plates are often obscured when the rear facing configuration is used. Hence for a site that is predominantly used by HGV's a front facing configuration would be chosen and conversely for a bus heavy site a rear facing camera configuration would be chosen. There is always a trade off between the vehicles captured using the front and rear facing cameras and the decision is made in the field and can be supplemented using the methods described in Section 3.3.4.

The Remote Sensing Device undergoes the same pre-measurement procedure as described in Section 3.3.3. The vehicle data collection process is automatic with the only input required from the user being to ensure that the alignment of the camera is optimal and minimise number of vehicle plates are being missed. Once configured the camera remains in the same alignment throughout the day unless other factors influence the image or alignment. Factors such as sun reflection can be removed by adjusting the exposure of the equipment. If the camera is moved inadvertently by passers by then the camera needs to be realigned. Throughout the course of the data collection process there was only one instance of a member of the public moving the camera.

The measurement process is automated by the control computer and requires no manual inputs. Information signs are located around the equipment to ensure that the public are aware that measurements are being taken, that we are a University study, that we are not able to prosecute them for speeding and that they should drive normally. It is important to inform the road users of this information so that the measurements are as representative of the driving environment as possible. It is also important to inform the road users because the deployment of cameras and equipment can induce erratic driving behaviour such as suddenly braking which not only decreases the validity of the measurement but puts unnecessary stress on the road users and others who may be following thereby increases the risk of an accident. Accidents and near misses induced by the placement of the instrumentation is highly undesirable as it not only limits the data collection but puts the drivers at risk and reduces the goodwill between the drivers and the monitoring crew. No instances of accidents were recorded throughout the data collection although one vehicle was observed to lock brakes through the instrument. After each observation, successful or otherwise, the database is automatically updated with the emissions measurement and the license plate photograph. Unsuccessful measurements are removed during post-processing

To ensure the efficacy of the measurements collected an audit procedure is required every two hours and is recommended every hour (see Section 3.3.3). For the audit procedure to be passed the Remote Sensing Device is required to provide three back to back measurements of the audit gas that fall within the experimental uncertainties built into the equipment. Failure to do this will result in the equipment being locked out with no further measurements being valid until a recalibration is performed and the audit procedure is passed. The audit process will ideally be performed when the road is free of traffic reducing the potential for transient background sources to influence the result. It is the responsibility of the operator to decide whether the failing of the audit is due to outside sources or the instrument requiring a recalibration and Section 4.2.2 discusses environmental factors that can influence the instrument measurement that are observable in the audit process. Under normal operating circumstances failure to meet the audit requirements on a regular basis will result in a recalibration being performed rather than trying to force a positive result.

The Remote Sensing Device will typically perform well on the audit procedure once initial calibration has been performed. The instrument is routinely re-calibrated at around mid-day on each day of measurements. This allows the data for the first half of the day to be backed up in case of electrical or system failure and is performed regardless of failure of the audit system. It also allows any major changes in environmental conditions that can influence the measurement of the instrument to be eliminated from the results.

After the observations are complete license plate data is then extracted by hand from the image by the instrument operator during post processing. Vehicle meta-data is sourced from Carweb (`http://www.carweb.co.uk`). There is a possibility that user error can be introduced here if the license plate is misread. To ensure the best possible quality of data entry any obscured or illegible plates are removed completely and any partial plates are cross-correlated with online resources that can check the model of vehicle so blatant errors such as confusing a car with a van can immediately be rectified. A number of vehicles were observed with partially obscured or illegible license plate sections. A freely available public license plate lookup website can be used to check for make and model for any given license plate (in this case `https://www.national.co.uk` was used). If the vehicle make and model is clearly identifiable then the publicly available information can presumptively identify a vehicle. For example if a Volkswagen Polo passed through the instrument with an unclear character in the image, for example R and A can be confused, one can be substituted in and if the resulting information states that the vehicle with that license plate is a Polo the missing letter can be confirmed. If the same test is performed with the result being a Ford Transit then that letter is discarded. This process is quick and efficient if internet access is readily available. If after exhaustively checking no reasonable match is made then that data is removed from the set as well. It is important to do the best job possible at this stage as no further quality control checks are performed on the data and if a license plate corresponding to a different car is entered at this stage there is no opportunity to remove it at a later date. Any results that are marginal are discarded and as such the few errors that might slip through at this stage ought not to influence the statistical methods applied to the data during analysis.

In addition to the emissions measurements the RSD systems fast operating light gates can calculate the speed and acceleration of the vehicle as it passes through. These measurements are validated using GPS in Section 4.2.1. Deployed at tyre level the instrument uses multiple gates with a fixed separation and calculates the acceleration based on the time differential. For wheel n breaking beam m at time $b_{n,m}$ and reconstituting the beam at time $r_{n,m}$ and where the beams have a constant separation s the speed (v) is calculated as $\frac{(b_{1,2}-b_{1,1})}{s}$ and $\frac{(r_{2,2}-r_{2,1})}{s}$. The time differential of these two measurements is used to calculate the acceleration (a).

Off Road Deployment

In this deployment, designed only to test the repeatability of the instrument (Section 4.2), a minimal configuration consisting of simply the Source Detector Module, the Corner Cube Mirror and the audit

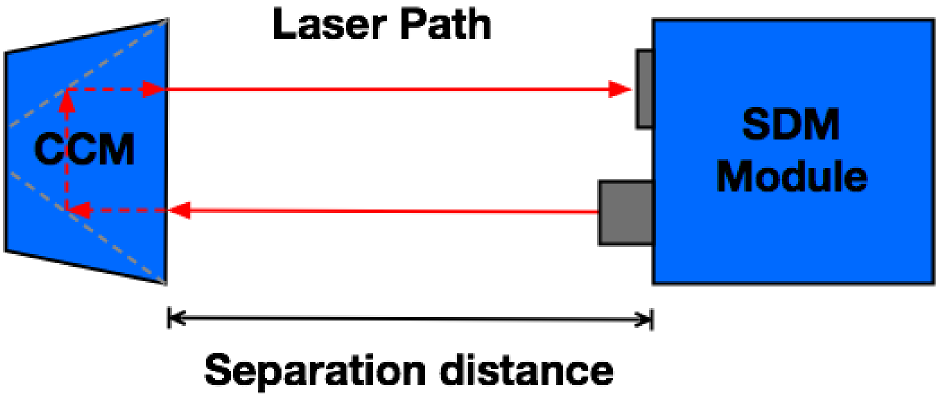

Figure 3.2: Schematic of the off-road experimental setup

gas canister was deployed for controlled gas audit tests as shown in Figure 3.2. The location is sheltered from the wind and a significant distance from public roads and with no through traffic the off road site is relatively free from sources of transient background interference allowing the most accurate baseline tests possible to be conducted. The site is highly desirable from a logistical point of view as it is within cable length of the mains power supply so no battery power is required. The site is also flat which means that the optics can be configured quickly and consistently.

Data is collected using repeated replications of the 'gas puff' audit method. A puff of gas with relative molecular abundances similar to that of a petrol vehicle exhaust plume is introduced into the beam path simulating a vehicle passing through. A measurement is taken in the same way as for a vehicle as described in Section 3.3.1. The process is used to audit the calibration of the equipment during on road experiments and are generally culled from the dataset during post processing however they can be extracted from the data set before post processing and used as measurements in their own right. The relative abundances of the control gas molecules are well known and fixed hence these measurements are the best way to test the accuracy of the instrument using repeated measurements. Vehicle exhaust plumes

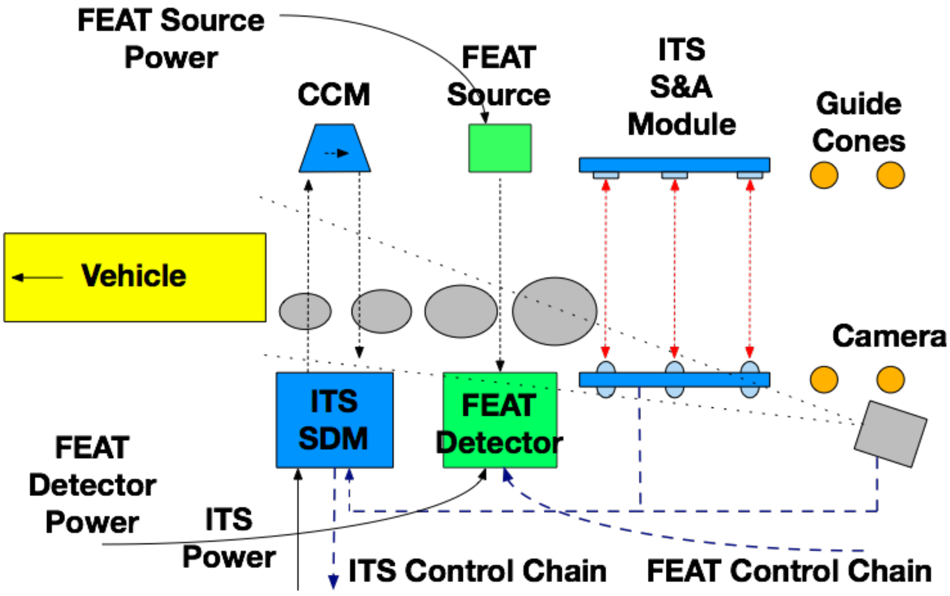

Figure 3.3: Schematic of the validation experimental setup

are subject to too many factors between ignition and emission to be reliable for repeated measurements useful in instrument validation.

Validation Deployment

The configuration of the ITS Remote Sensing Device is the same as for the standard on-road deployment 3.3.3 with the addition of the FEAT system provided by The University of Denver. The FEAT system is co-located with the RSD4600 in a closed car park in Leeds as shown in Figure 3.3. The validation experiment took place over 8 days so to ensure consistency the locations of the instruments were photographed and marked.

Vehicles were made available by the University of Leeds, Leeds City Council and by members of the public. These vehicles were driven through the equipment as if they were driving on a road. Vehicles were categorised by the gear they were driving in and whether or not they were accelerating through or cruising. Accelerating vehicles were required to stop at the guide cones and accelerate through the equipment and cruising vehicles were driven through from a rolling start at a comfortable engine speed as determined by the driver. Limitations on space meant that gears beyond third were impossible as the vehicle was unable to pick up enough speed to stop the engine from stalling.

Post processing of the data required the vehicle license plates to be captured from the camera and entered into a database. The meta-data was then requested from the Carweb database in the same way as for on-road measurements. As all vehicle license plates were known prior to them completing their runs and all the time data was logged there was no ambiguity in any of the vehicles and all valid measurements were assigned their correct license plate. The two RSDs had a small timestamp offset and this was identified and corrected during the measurement process. The measurements across the two instruments were aligned using the measurement timestamps.

There were two significant issues identified during this measurement process. Firstly the presence of dust in the car park meant that the selection of the location was important. Initial locations resulted in too much dust being picked up and re-suspended in the wake of the vehicles and this dust caused too much obscuration of the beam to give valid results. The solution to this was to move the equipment to a set of concrete slabs. The dust still presented a problem although measurements could still be taken. Vehicles would pick up dust when off the slabs and deposit it there when they drove through the equipment. Regular sweeping of the area used for measurement provided some reduction of dust based obscuration however some measurements were still missed because of this issue. In addition to the resuspended dust the generators used to power the FEAT system were initially a source of transient background pollution that meant the audit procedures regularly failed. Initially the source was thought to be due to a railway line a few hundred meters upwind the site but once the generators were identified as a likely source changing the position of the generators to somewhere downwind of the RSD immediately resolved this problem.

3.3.4 Additional Measurement Methods

A number of other methods were developed and used throughout the course of the three years of data collection to account for the compromises in the RSD measurement procedure identified earlier. Initially, starting in 2014 at Leeds (Section 3.4.3) and continued until the beginning of the Aberdeen Study (Section 3.4.3), an additional operator was seated somewhere with a clear sight line to the vehicles and given a camera. Every time a vehicle that might not be captured by the static camera was identified a photo was taken of the front of the vehicle capturing the license plate. With busses the operator identification number, visible on the front back and side of the vehicle, is recorded and correlated with the license plate and where possible the photograph can be used to identify the license plate for the vehicle. For HGV's the process is more involved as they do not have the same additional identification numbers that busses do. Photographs are taken as for busses however the time stamp on the image and the characteristics of the vehicle in both photographs are analysed and if the secondary picture can be used to identify the vehicle in the first photograph. Whilst initially convoluted an experienced analyst can identify most HGV's based on a combination of colour and time. After this careful checking process vehicles with a license plate that remains unmatched or where there is uncertainty are discarded.

There are a number of limitations with the manual acquisition of photographs of license plates. The resolution of the camera often leaves illegible results, reflections and other interference can also pose a problem and the limitations on the field of view mean that a license plate might be missed completely. To solve this problem a GoPro^{TM} camera was used to measure a constant video stream of passing vehicles. This method was first trialled on Day 1 of the Aberdeen study in April 2015 (Section 3.4.4) and successfully used for the rest of the study. The GoPro captures video at 720p resolution which is enough to capture all but the most obscured license plates, where dust mud and other unwanted species completely cover the plate, or those offset outside of the field of view. This method eliminates the need to constantly be monitoring traffic and provides multiple opportunities to visually correlate vehicles captured by the license plate camera with their license plates obscured.

The GoPro solution has a number of drawbacks. The camera batteries need to be replaced every 2 hours and the data collected requires a large amount of memory to store it compared to the images taken with the RSD camera. Additional memory was made available through the use of a high capacity portable hard disk drive. GoPro cameras are by design light and portable and therefore the risk of theft is high. To combat this the GoPro was mounted in a traffic cone with a section removed creating a viewfinder and secured using a combination of cable ties and tape making it only removable by the most determined and well prepared individuals. The data requires a lot of time to process. Monitoring of the time of the measurement and the file save time can provide a ball-park figure however ultimately identifying the vehicle requires watching large sections of the video stream. This method is highly effective at identifying vehicles that would otherwise be lost and can be used for additional purposes such as mapping the total fleet including those that did not provide valid gas measurements for the Remote Sensing Device.

3.4 Site Descriptions

As previously alluded a wide range of sites in Sheffield, Cambridge, Aberdeen and Leeds were observed throughout the thesis. The following section details the observation projects that have been used in this thesis in chronological order. Where possible photographs and maps are presented for each site. Figure 3.4 shows a three panel plot with the locations of all fifteen primary on road observation sites marked along with their name which will be used to refer to them throughout the rest of this section.

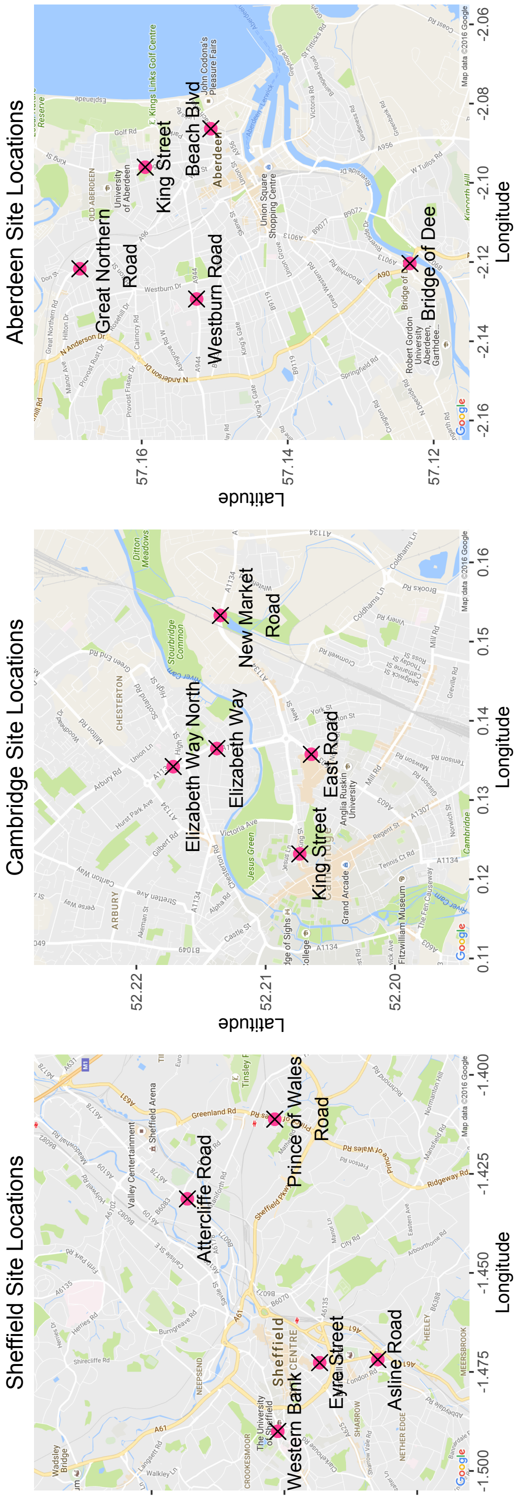

Figure 3.4: Locations of main on-road observation sites

3.4.1 Sheffield Data Collection Project

Introduction

The University of Leeds through the Institute for Transport Studies conducted a 10 day observation project in Sheffield (West Yorkshire, UK) during the summer of 2013 for the local council as part of a feasibility study into a low emission zone in Sheffield. 10 days of measurements, 2 each at 5 different sites around Sheffield were conducted. The results and policy recommendations have been submitted to the council Tate (2013b). The data collected in Sheffield was retained by the University of Leeds. Further analysis on this data set, henceforth referred to as the *Sheffield Data Set*, was undertaken as part of this thesis.

Site Selection

In accordance with the site selection guidelines described in Section 3.3.3 five sites were selected for measurements. The sites represent a range of different road types and locations around Sheffield. The sites selected were as follows (presented in alphabetical order):

- Asline Road
- Attercliffe Centre (A6178)
- Eyre Street
- Prince of Wales Road (A6102)
- Western Bank (A57)

Asline Road

Asline Road is a road located in the South of Sheffield. It is on the outskirts of the city centre and has a relatively low traffic flow. It has an elevation of 74m and a road gradient of 1.1°. Data was collected from Asline Road on the 23 and 24 April 2013. Data was collected from

Figure 3.5: Asline Road site photograph

1026 - 1800 and 0818 - 1800 with 1156 and 1675 valid measurements on each day respectively. Asline Road is a relatively quiet site compared to other Sheffield sites and was the first site observed in this thesis. Whilst live data was collected over both observation days this site was also used as a training opportunity to learn how to set up the Remote Sensing Device in real world conditions.

The Asline Road site was also used to test an experimental body sensor module to aid in the capture of HGV license plates. The tests showed that the body sensor module was not suitable for use in this experiment however it could be used in future experiments alongside a scaffolding structure to measure the emissions from elevated exhaust pipes.

Attercliffe Centre (A6178)

The Attercliffe Centre site is located to the North of Sheffield in a prominent industrial location. It is located at an altitude of 54m with a road gradient of 1.7°. The road connects to the centre of the city and has a high traffic flow including a large number of HGVs. There is little service by public transport resulting in very few buses observed. Data was collected from Attercliffe Centre on the 4 and

Figure 3.6: Attercliffe Centre installation

5 June 2013. Data was collected from 0802 - 1800 and 0812 - 1800 with 4180 and 4594 valid measurements on each day respectively. The traffic flow is considerably higher than Asline Road and has more total measurements than any other site in Sheffield.

Due to the low volume of busses passing this site and the availability of required space on the roadside a decision was made to re-orientate the license plate to the forward facing configuration (see Section 3.3.3 for description and Figure 3.6 for example in situ) meaning that the license plate capture of HGV's with ground level exhausts was higher than at other sites without the inevitable loss of bus data that comes with this configuration.

Eyre Street

Eyre Street is located in the city centre. It is at an elevation of 70m and has a flat gradient of 0°. The site is located next to a public market and is well served by busses. Data was collected on the 30

Figure 3.7: Eyre Street installation

April 2013 and the 1 May 2013. Data was collected from 0814 - 1801 and 0800 - 1800 with 2500 and 2460 valid measurements collected on each day respectively. The site is a typical inner urban site in terms of traffic flow. There are few HGVs using this section of the road so the license plate capture camera can be deployed in the rear facing configuration without risking significant loss of measurements from this vehicle class. The high number of busses present at this site also means that the rear orientation of the camera makes the capture rate of these vehicles considerably higher than could have been achieved with the front facing camera.

Prince of Wales Road

Prince of Wales Road is located to the East of the city in a similar geographical area to Attercliffe Centre (Section 3.4.1). The site is located on a section of outer ring road. The site is located at an elevation of 74m and has a road gradient of 1.8°. Data was collected

Figure 3.8: Prince of Wales Road installation

on 12 and 13 May 2013. Data was collected from 0820 to 1637 and 0825 to 1327 with 4064 and 1373 valid measurement collected on each day respectively. The site is heavily used with a mixture of vehicles of all types using the road.

Both days but especially the second day was interrupted by poor weather. The Remote Sensing Device was designed in Arizona and is not waterproof. Weather conditions at one point were so poor that the Remote Sensing Device had to be removed from the roadside or it would have been washed away. Nonetheless a large number of measurements were taken and the time spent at the site was not wasted.

Western Bank

Western Bank is located near the University of Sheffield and the site was chosen next to the local children's hospital. A site of particular importance given that children are at additional high risk from air pollution (Kulkarni & Grigg, 2008; Raysoni & Li, 2009). The site is located at an elevation of 131m and has a gradient of 2.3°. Data

Figure 3.9: Western Bank installation

was collected on 13 and 14 May 2013. Data was collected from 0824 to 1800 and 0830 to 1800 with 3121 and 3084 valid measurements collected on each day respectively. The site is well used with a wide range of vehicles including high proportion of busses.

The space available at the site meant that it was impossible to arrange the camera in the ideal rear facing configuration and a front facing configuration in a similar fashion to the Attercliffe Centre site. Busses were identified using their bus identification number as opposed to their license plate. License plate to bus identification number correlation was taken from either the Eyre Street images where both are clearly visible or from the rarer occasion when both are visible on the Western Bank images. The only available images were those from the installation camera as this data was collected before experimenting with other ways to collect missing license plate data as described in Section 3.3.4. When identification numbers were visible but the license plate was obscured the reference table was consulted and the appropriate license plate was added. Where no correlation was possible the measurement was discarded.

3.4.2 Cambridge Data Collection Project

Introduction

The University of Leeds through the Institute for Transport Studies conducted a 10 day observation project in Cambridge (Cambridgeshire, UK) during the summer of 2013 for the local council as part of a study to investigate ways to reduce the air pollution levels in the city. Ten days of measurements at five different sites, two days at each, were conducted between the 7 May 2013 and 7 June 2013. The results and policy recommendations have been submitted to the council (Tate, 2013a).

Site Selection

In accordance with the site selection guidelines described in Section 3.3.3 five sites were selected for deployment of the Remote Sensing Device. The sites represent a range of different road types and locations around Cambridge. The sites selected were as follows (presented in alphabetical order) and can be seen in the middle panel of Figure 3.4.

- East Road
- Elizabeth Way (North: A1134)
- Elizabeth Way (A1134)
- King Street
- New Market Road

East Road

East Road is located close to the shopping centre of Cambridge but away from the historical centre and has the highest traffic flow of all sites measured. The site has an elevation of $17m$ and a gradient of $-0.1°$. Data was collected on 21 and 22 May 2015 with from 1150 -

Figure 3.10: East Road installation

Figure 3.11: Elizabeth Way installation

1800 and 0802 - 1800 respectively. The loss of recording time on the 21 May was due to weather conditions being unsuitable. The total number of valid measurements was 1833 and 3046 for the 21 and 22 May respectively.

Elizabeth Way North and Elizabeth Way

Two sites were selected on Elizabeth way. One site was located North of Cambridge in a suburban location (Elizabeth Way North)

Figure 3.12: King Street installation

and another was located more centrally (Elizabeth Way). Elizabeth Way North is positioned on the exit of a roundabout and Elizabeth way is located more centrally and is shown in Figure 3.11. Elizabeth Way North and Elizabeth Way are located at an altitude of 10m with a gradient of 0.6° degrees and 1° respectively. Measurements for Elizabeth Way North were taken on 7 and 20 May 2013 from from 1113 - 1800 and 1118 - 1800 and for Elizabeth Way North and 23 May and 6 June for Elizabeth Way from 0845 - 1349 and 0816 - 1800 respectively. The total number of valid measurements from each site were 1272, 983, 1076 and 3118 respectively. Both sites were conveniently located with good traffic flow. The loss of survey time was due to inclement weather conditions.

King Street

King Street is located in the centre of the historic district of Cambridge with the measurement site located on the opposite side of the

Figure 3.13: New Market Road installation

road from Jesus college. King Street is located at an altitude of $14m$ and has a gradient of $-0.2°$. Data was collected on the 8 and 9 May 2015. Data was collected from 0949 - 1800 and 0815 - 1533 respectively. The time lost on both days was due to poor weather conditions. The total number of measurements was 394 and 291 for 8 and 9 May respectively.

King Street is a narrow road with traffic flowing in both directions. Vehicles that were traveling in the opposite direction to the configuration of the RSD were discarded. Figure 3.12 shows a vehicle passing through the measurement site in the correct direction. The traffic flow at this site was not ideal with more measurements possible at all the other sites however it is representative of sections of the road network in Cambridge so the negatives due to low flow rate and low number of measurements are offset by the increase in completeness of network conditions measured.

New Market Road

New Market Road is located close to the Cambridge United football stadium and has a high traffic flow rate compared to other sites. The site has an elevation of $11m$ and a gradient of $2.6°$ degrees. The gradient at this site is the greatest encountered during the Cambridge measurement program as the site is located on the approach to a bridge. Cambridge sites are notoriously flat. Data was collected on 10 May and 7 June from 1031 - 1800 and 0818 - 1800 respectively with lost measurements due to setup time on the 7 June. The total number of valid measurements was 1497 and 1881 for the 10 May and 7 June respectively.

The site was set up in the standard configuration with the remote sensing device and vehicle located in the central reservation as shown in Figure 3.13. During the morning peak session the width of the road allowed vehicles to go through side by side when congested. These results were unsuitable for measurement as the RSD is unable to distinguish the individual plumes from separate vehicles. The measurements with multiple vehicles caught in the camera are therefore discarded as invalid.

3.4.3 Leeds Study

A number of days of study were performed at a site in Leeds. The site can be seen in plan view in Figure 3.14 courtesy of the Google Maps satellite. A number of different projects were undertaken at the Leeds site but the primary reason for using this site as part of this thesis is for the testing of various different methodologies for removing the lost data due to the issues mentioned in Section 3.2.3 and are described in Section 3.3.4.

Figure 3.14: Satellite view of the Leeds installation (Google^{TM})

3.4.4 Aberdeen Study

Introduction

The University of Leeds through the Institute for Transport Studies conducted a ten day observation project in Aberdeen, Scotland's third most populous city, during the summer of 2015 as part of a feasibility study for a Low Emission Zone in the city centre. The results of this study are published as part of the annual report by Aberdeen City Council (Tate, 2016).

As part of this thesis the data collected in Aberdeen was retained by the University of Leeds. Further analysis on this data set, henceforth referred to as the *Aberdeen Data Set* was undertaken as part of this thesis. The data from Aberdeen is of particular importance as it is the first to contain data collected after the Euro 6 vehicle emissions legislation begin to come into force. Despite the presence of Euro 6 vehicles in both the Sheffield and Cambridge data sets, their numbers were low and it is difficult to make any statistically sound statements about. The Aberdeen data set will overcome start to overcome this issue and allow us to better understand the impact that Euro 6 legislation has had on real driving emissions.

A significant problem was discovered half way through the measurement campaign. A power supply unit used for charging the batteries powering the equipment over night was found to have a faulty connection and was failing to charge the batteries overnight. Initially this was thought to be down to user error but as the problem persisted it became clear that the issue was with the power supply. After thorough investigation the issue was fixed however there was significant data loss at a number of sites throughout the second week.

Site Selection

A number potential sites were chosen for monitoring prior to arrival by the measurement team. After further investigation upon arrival the number of sites was reduced to five based on the logistical requirements of the sites. The five sites chosen were

- Beach Boulevard
- Bridge of Dee
- Great Northern Road
- King Street
- Westburn Road

Beach Boulevard

Beach Boulevard is a road located East of Aberdeen City Centre and is one of the main link roads to the amusement arcade and beach front area of the city. It is approximately at sea level and has a road gradient of xx degrees. Data was collected from Beach Boulevard on 18 and 24 May 2015. Data was collected from 0755 - 1200 on the 24

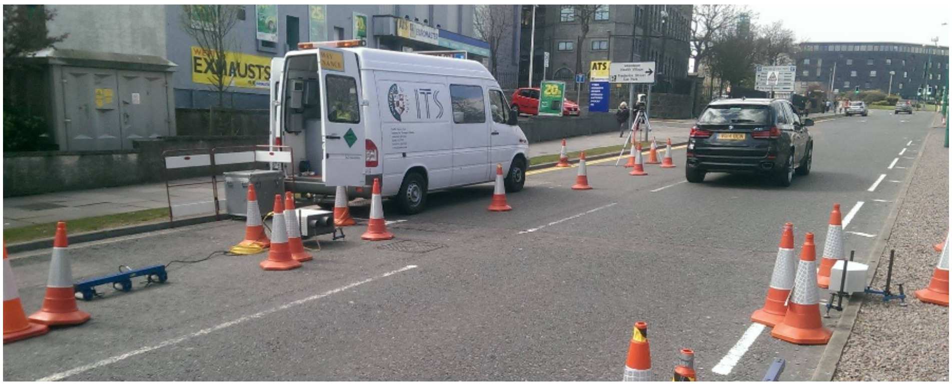

Figure 3.15: Beach Boulevard Site photograph

May and no valid data was collected on the 18 May. The total number of valid measurements was 593.

Based on site scouting it was anticipated that Beach Boulevard was not likely to contribute a significant number of measurements to the overall total due to the low vehicle flow and was used as a site for days where it was anticipated that the number of measurements would be low regardless of the site selected to mitigate this negative impact. The first day at the site was a Saturday, typically not a day monitored during remote sensing studies. In the Aberdeen study observations were made on Saturday to allow time to drive back to and from Leeds. Furthermore this site was hit by a large scale power failure caused by a malfunction of one of the battery charging units (described fully in Section 3.4.4) causing all data to be lost during the first day. Beach Boulevard was also used as the final day of measurements on the 24 May. The site was intentionally shut down at mid-day to allow a safe return drive to Leeds immediately following it. As the site was monitored at anticipated low traffic days the number of observations lost was minimised. Under normal conditions this site would not have been selected as an observation point however due to restricted access for van parking and safety issues of connecting the cables to the instrument at the other potential sites Beach Boulevard was the last viable

Figure 3.16: Bridge of Dee Site photograph

site that could be used. In the context of this thesis another day at a higher traffic flow site would have been more desirable for increasing the number of observations however it was important for the survey to make measurements at a wide range of observation sites so Beach Boulevard was chosen.

Bridge of Dee

The Bridge of Dee is a choke point for vehicles leaving Aberdeen to the south. At this location the road network is almost always running at full capacity from the start of the day until the end. The site is located immediately after a junction at approximately sea level. Data was collected on 16 and 17 May 2015 from 0846 - 1800 and 0804 - 1800 on respective days. The total number of valid measurements was 6686 and 6933 on the 16 and 17 May respectively. The site was configured with a front facing camera despite there being no HGV's using the road as there was not space to mount the camera in the rear facing configuration.

Time was lost on the first day as access to the site was compromised and calibration was not performed in time for the scheduled 0800 start. The biggest cause for concern at the Bridge of Dee was that the constant flow of at-capacity traffic meant that it was difficult

Figure 3.17: Great Northern Road Site photograph

for the instrument to be audited effectively. The solution used was to use the pedestrian crossing to force breaks in the traffic and allowing a window of clear road to perform the audit. This process was hindered by the presence of transient NO_X sources from passing traffic on the other side of the road that were a source of contamination for the measurements. The standard audit procedure typically takes 5 minutes to perform however at this site it could take upwards of 1 hour to perform. The window for auditing is ideally 1 hour but the equipment allows for a 2 hour window meaning that the audit process was always completed in time.

Great Northern Road

Great Northern Road is located to the North of Aberdeen and is located immediately after a roundabout before the traffic becomes dual laned. Data was collected on the 22 and 23 May 2015. Data was collected from 0800 - 1208 and 0800 - 1700 respectively. The total number of valid measurements was 592 and 841 for the 22 and 23 May respectively. Data was lost due to power failure caused by charging issues described previously. The site was configured using the front facing camera as there was not enough space to have it facing forward.

Figure 3.18: King Street Site photograph

King Street

The King Street site was the first site used in the Aberdeen study as it was located close to where the operators were staying. It is located close to Aberdeen University and is used by both HGV's and Busses regularly. Data was collected on the 14 and 15 May 2015 from 0814 - 1800 and 0827 - 1801 respectively. The total number of valid measurements was 2958 and 2729 on the 14 and 15 May respectively.

The road way was particularly narrow at the monitoring station as shown in Figure 3.18 meaning that larger vehicles had to slow down significantly to pass through the instrumentation safely. This presented a problem as it is not as representative of real world driving as some of the other sites however it was the best place to record HGV's entering Aberdeen. The King Street site was the first use of the cone-cam GoPro installation (see Section 3.3.4) and the results from the cone-cam, whilst incomplete as the users familiarised themselves with the method, could immediately be used to pick up HGV's that had not had their license plates captured by the standard camera.

Figure 3.19: Westburn Road Site photograph

Westburn Road

Westburn Road is located to the west of Aberdeen in a suburban environment. The road is a tributary road to the outer ringroad. Data was collected on the 20 and 21 May 2015 from 0817 - 1800 and 0816 - 1700 respectively with time lost due to power failure. The total number of valid measurements were 2487 and 2465 for the 20 and 21 May respectively.

Morning access to the site was often limited by the presence of peoples' private vehicles blocking sight lines for the cameras or being parked in reserved bays. There was no way around this except to try and pick locations that mitigated these issues as much as possible. The Westburn Road monitoring took place towards the end of the fortnight and the power failure issue was becoming predictable. Measures to limit the impact of it were able to be taken in advance of the problems happening so more data was able to be saved than from sites where the issue was not fully understood.

3.4.5 Zurich Data Set

Whilst not measured as part of this PhD a data set from a remote sensing team operating from Zurich was made available for analysis by the original collectors. This dataset, the Zurich dataset, was collected between 11 June 2014 and 28 August 2014 as part of a long term study at the same site from the year 2000 (Chen & Borken-Kleefeld, 2014). The measurement site for this dataset was located at a bus stop intersecting a junction serving a commercial and school area. The gradient at the site is 9.2°. The site is preceded by a $\approx 1.5km$ uphill drive ensuring that as many of the engines as possible are running hot. A total of 36985 measurements were made available from this dataset. Metadata is also available on the make, model, fuel type and euro standard although not as comprehensive as the RSD data collected as part of this thesis. This dataset is used in conjunction with the other data detailed in this chapter to primarily assess the effectiveness of Euro 6 vehicles in Sections 5.2.1 and 6.2.1 as it represents a relatively clean set of vehicles that have been exposed to a similar driving environment immediately prior to measurement.

A benefit of this location is that the engines are observed after hot running. It is very unlikely that any vehicle that is observed at this site is operating under cold start conditions so it is a reasonable assumption to exclude this effect from any measured observations. The site has a good traffic flow with approximately 5000 vehicles observed per day, comparable to the higher traffic flow sites observed in the Aberdeen, Cambridge and Sheffield observations. A wide range of Euro class and fuel types are available for analysis from this dataset and it provides an ideal compare-and-contrast style data set for the urban sets used throughout the thesis.

3.5 Summary of Data

Over 6×10^4 valid measurements were collected over the three main observation projects undertaken during the writing of this thesis and are summarised in Table 3.5. A range of different sites were monitored with both relatively steep and flat gradients encountered. The difference in sites measured means that comparisons between different sites can be presented in a meaningful way. Most importantly 782 Euro

6 passenger vehicles were observed in Aberdeen and a small number were observed in other surveys. The Euro 6 measurements are critical to the development of the goals of the thesis. Additional remote sensing data is also available from a site located close to an off-ramp in Switzerland which was not collected as part of this research project is described fully in it's own publication (Chen & Borken-Kleefeld, 2014).

Location	Car	Taxi	LCV	HCV	Bus	Coach	Total
Aberdeen	19120	496	3500	515	273	120	24024
King Street							4900
Bridge of Dee							13129
Beach Boulevard							569
Westburn Road							2478
Great North Road							2960
Cambridge	11633	643	2408	253	454	-	15391
Elizabeth Way (North)							2255
King Street							685
New Market Road							3378
East Road							4879
Elizabeth Way							4194
Sheffield	19978	1068	4808	691	1654	8	28207
Asline Road							2831
Eyre Street							4960
Western Bank							6205
Attercliffe Centre							8774
Prince of Wales Road							5473

Table 3.1: Summary of data collected across the three main observation sites

The number of vehicles and the different types of vehicles observed present a wealth of data only available in remote sensing based studies. A key differentiator of the remote sensing methodology is that it begins to use a *big data* approach compared to a much more sampled approach favoured by PEMS and by laboratory based studies (Section 2.5). The big data approach means measuring lots and lots of different vehicles so that the behaviour of the fleet as a whole is what is being observed. In doing this any vehicle that is behaving atypically of the fleet can be identified in post process. The big data approach means that if factors such as catalyst degradation due to poisoning or physical damage and cold start effects, if significant in the fleet, will be observable in this data set. These vehicles would likely present themselves by exhibiting off-model characteristics compared to the rest of the fleet vehicles in a given subset. Attempts to model the fleet are described in Chapter 5 and forward throughout the rest of the thesis.

Chapter 4

Validation of Equipment

4.1 Introduction

This chapter presents the testing of the Remote Sensing Device and ancillary equipment to prove beyond all reasonable doubt that all the measurements are robust. This is done to bolster confidence in the methodology and further prove that the results obtained using an RSD can be used as reliable vehicle emissions evidence. It also aims to illustrate the limitations of the instrumentation and identify potential pitfalls that need to be accounted for. The RSD used throughout the majority of this thesis does not have the ability to directly measure NO_2, a key pollutant in vehicle emissions and so this chapter also attempts to relate the NO_2 measurements taken using a state of the art research prototype RSD to the NO measurements taken by the RSD4600. The aim of this is to improve the estimates of NO_X emission made by the RSD4600 in the absence of any other way to measure NO_2 directly, improving the evidence from the ITS instrument (RSD4600) allowing estimates of total NO_X to be improved in future chapters and thereby addressing one of the key research gaps identified in the literature review. The results presented in this chapter are an extended version of a paper under review by the Journal of Air and Waste Management.

4.2 Validation of RSD4600 Equipment

A number of experiments were performed off road to test the basic running of the remote sensing equipment and it's responses to changes in ambient conditions. The speed and acceleration module was tested against industry standard GPS technology and the Source and Detector Module (SDM) was rigorously tested to validate the measurements presented throughout the rest of this thesis.

4.2.1 Accuracy of Speed and Acceleration Measurements

The speed and acceleration measurements described in Section 3.3.3 taken by the RSD are used to calculate the VSP of each vehicle. This is an important diagnostic tool and is used in Section 5.3.2 and Carslaw *et al.* (2013) amongst other studies to better understand the distribution and cause of the most highly emitting vehicles. The accuracy of the speed and acceleration measurements is critical to the conclusions drawn from any VSP based study. To test the accuracy of the RSD4600 speed and acceleration module measurements were taken over eight days at an off road car park location in South Leeds and compared to results gathered simultaneously using a state of the art onboard GPS system. A number of different vehicles and driving conditions were tested allowing for a large range of speed, acceleration and hence vehicle specific power to be tested. These measurements are representative of most realistic driving conditions encountered by vehicles driving in an urban environment. The range of vehicles tested was extensive from single deck passenger busses fitted with automatic transmissions to small passenger cars more representative of the majority of fleet vehicles. A number of light to medium commercial vehicles were tested as well.

As discussed in Chapter 2 vehicle specific power is increasingly being used in vehicle emissions studies. The advantages of the RSD4600 speed and acceleration module is it is fast acting and portable with a low measurement refusal rate. Validation of this methodology has been performed using a state of the art global positioning system (GPS) provided by VBOX Automotive Systems and used in previous studies (Wyatt *et al.*, 2014). The GPS has a circular error probability of $3m$ latitude and longitude and has a sampling rate of $10Hz$ (VBOX, 2008). The remote sensing equipment was set up as shown in Figure 3.3. Vehicle fitted with the GPS system were repeatedly driven through the equipment and both the RSD and GPS speed and acceleration was recorded. The data reduction process required a number of steps. Firstly the clock times needed to be synchronised. This was performed by observing the two measurements' time series of results and aligning them based on the spaces between the RSD measurements. An offset of 26 seconds between the clock times of the RSD and the GPS was observed and adjusted for. This process is non-trivial as the repeated nature of the measurements makes one look very much like another. By carefully comparing start and stop times as well as speed profiles it was possible to become confident with the matching of the GPS time stamps to the RSD time stamps.

A data selection window is also specified based on the GPS location. To select the data the latitude and longitude coordinates are transformed into radial polar coordinates with the origin set as the centre of the measurement area. This is necessary because the alignment of the equipment basis is not aligned with the basis of latitude and longitude. The selection window overlaid with the polar coordinate basis set is shown alongside the data that was selected in Figure 4.1. The selection window and the schematic representation of it are not to scale. The oval shape of the data rather than the circle shape of the schematic is because of the difference in scales between the latitude and longitude axes on the underlying graph. All GPS points that

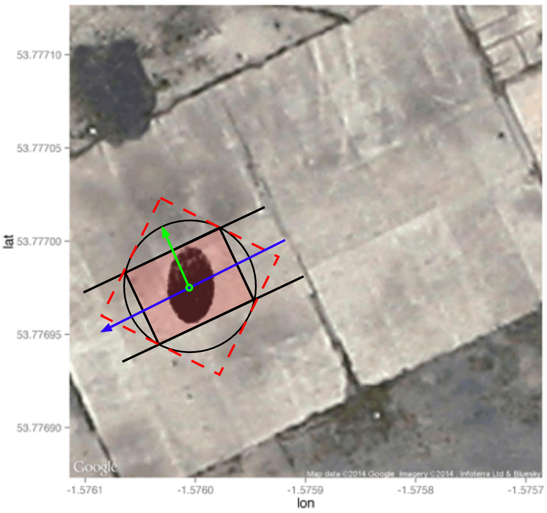

Figure 4.1: Schematic of polar coordinate transformation

fell within this window were selected. To ensure no further unwanted measurements were included through GPS measurement error a further check against the two instruments' time stamp was performed. A 5 second window was used for further exclusion of GPS data that did not represent the RSD4600 data but might have been selected due to errors in GPS. Through this double-lock selection criteria simultaneous measurements of the wheel speed through the light gate and the GPS were taken for a range of different vehicle speeds and accelerations allowing the RSD4600 speed and acceleration values to be validated against industry leading technology.

In total 337927 GPS points were recorded including the period of stopping and starting the equipment and driving to and from the RSD. GPS data that met the selection criteria were selected for analysis with 1894 falling within the relevant selection window (0.56%). The selection window represents a very small portion of the loop driven for each pass through and the small percentage selected for analysis. From these points a further 186 GPS points (9.8%) removed due to

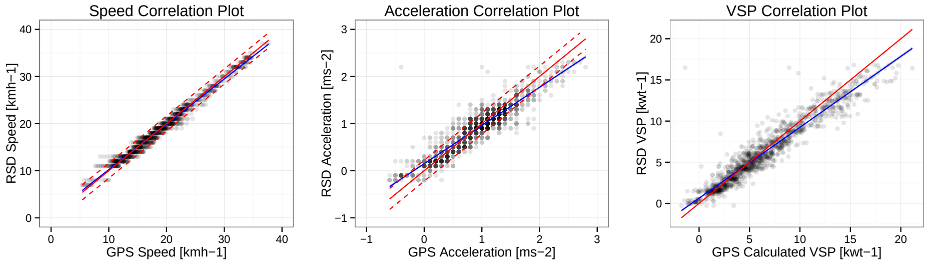

Figure 4.2: Speed, Acceleration and VSP GPS to RSD correlation plots.

driver error leaving a total of 1708 GPS points left for analysis. These points were matched with 328 valid speed and acceleration measurements from the RSD. The selected speed and acceleration data were plotted in Figure 4.2. The VSP was calculated using Equation 4.1 for speed in miles per hour and acceleration in miles per hour per second and plotted alongside. A linear model of the form $y = c_1x + c_2$ was fitted to the speed and acceleration and a 95% confidence interval was calculated using the fitting algorithm. The coefficients for the speed and acceleration models are $c_2 = 0.61$ and 0.47, $c_1 = 0.96$ and 0.83 and adjusted R^2 values of 0.98 and 0.86 respectively. The linear model for the VSP has $c_2 = 0.52$ and $c_1 = 0.88$ and an adjusted R^2 value of 0.92.

$$VSP = (0.2va)+(4.39vsin(\theta))+(95.4\times10^{-3}v)+(27.2\times10^{-5}v^3) \quad (4.1)$$

The results show a strong positive correlation between the GPS measured speed and acceleration and the RSD4600 speed and acceleration module results and hence the derived VSP values. The vast majority of results fall within the error of the instrument. The 95% confidence interval derived from the linear model of the data is within the error for the equipment(±1 mph). Acceleration requires more steps to measure and as such the measurement error is greater than that of velocity ($\pm 0.5mphs^{-1}$) however the RSD4600 acceleration measurements still correlate very well with the GPS measurements. There is some evidence that the RSD underestimates the acceleration at the most extreme values however those levels of acceleration are not typical of real world driving and are still largely within the error of the instrumentation. It is concluded that the speed and acceleration measured by the RSD4600 is validated and therefore the VSP calculated is also validated.

4.2.2 Temperature Sensitivity of the Equipment

The RSD instrument requires 45 minutes at the start of a session to reach a thermal equilibrium. This means that the impact of ambient temperature on any systematic error must not be overlooked. Whilst the remote sensing device measurement days are typically performed under good weather conditions the ambient temperature can vary quite significantly between the start of the day at 0800 and shutting the instrument down at 1800, a typical time window of operation. A variation of $10°C$ or more can be observed and exposure to direct sunlight can also influence the instrument temperature. To understand the relationship the equipment was tested throughout the course of one day in an off road area. The equipment was calibrated initially at 0934 and the first measurement was performed at 0937. Measurements were taken at regular intervals throughout the day using the 'gas puff' audit method described in Section 3.3.3 and the final measurement was taken at 1600 with 339 measurements being taken in total. Along with measuring the temperature variance over a day another aim was to empty the gas bottle so fewer measurements were taken at the start of the day to ensure that gas was available at the end. More measurements were taken later when it became clear that there was more gas remaining in the bottle than anticipated. Along with the NO the instrument also records the ambient temperature.

No instrument calibrations are performed between the initial and final measurements and no adjustments were made to anything else. The result shows only variation in response to changes in ambient conditions. The results of these measurements are shown in Figure 4.3. The initial temperature and the NO concentration in the gas bottle are also shown on their respective graphs as horizontal lines.

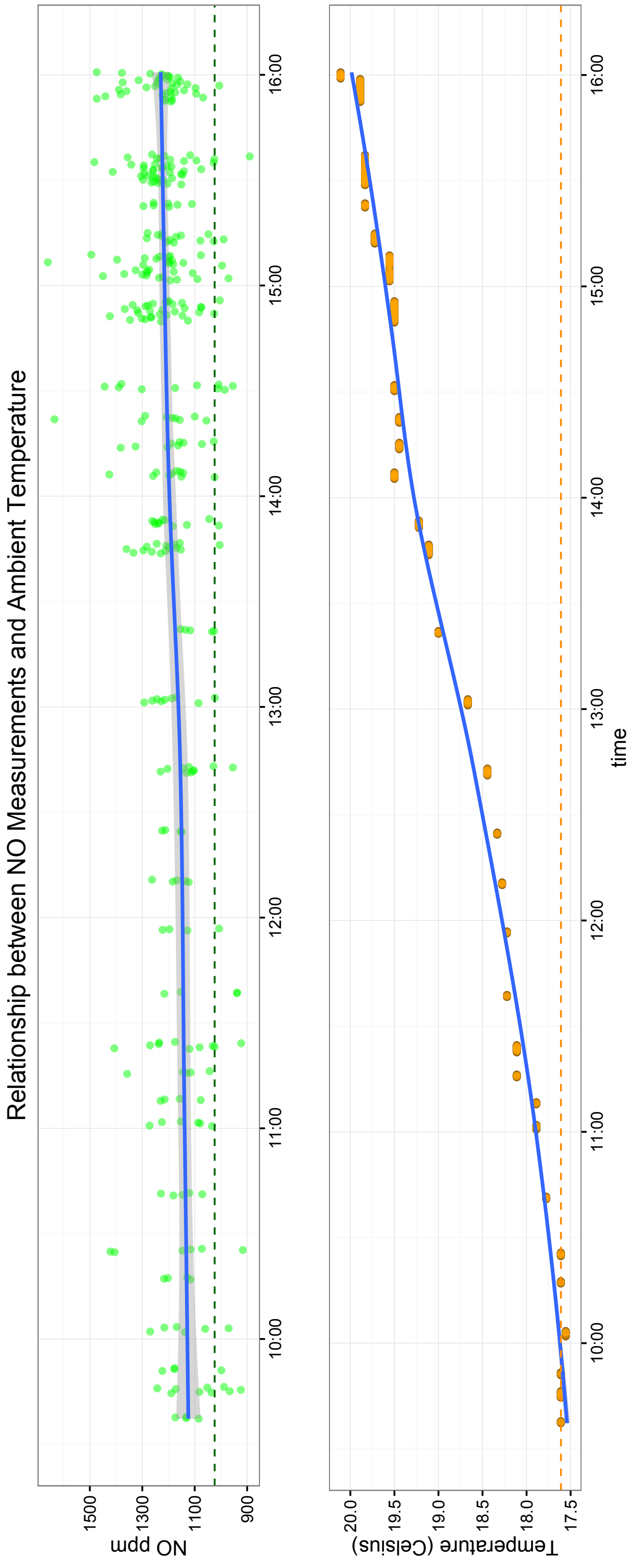

Figure 4.3: Time series plot of both the NO and the temperature.

The variation in temperature over the course of the day is $2.5°C$, significantly smaller than the maximum variance observed on some observation days although the start and finish times are both later and earlier than normal respectively. Despite the small change in temperature a variation in NO measurements as well as a change in trend that is generally in line with the trend of the temperature is also observed. The trend line is 100ppm greater in the afternoon than in the morning. 100ppm is 10% of the measurement and is a significant result. The change in the observed measurement was enough to trigger a recalibration under normal operating circumstances as the instrument would have repeatedly failed to pass the audit procedure. This result shows that the change in ambient conditions should not be considered as an afterthought and any operator of the remote sensing device needs to be aware of changes in ambient conditions whilst conducting measurements. The AM and PM calibrations should be seen as the absolute minimum that should be performed and any change in local weather, for example the cloud cover changing, should be accompanied by a recalibration of the equipment. This experiment was performed prior to the Aberdeen survey and over this survey the instrument was re-calibrated whenever significant changes in ambient conditions were observed as well as the standard AM / PM re-calibration.

4.2.3 Distance Sensitivity of the Equipment

Often the total path length of the laser can change from site to site and in smaller amounts from day to day. It is important to understand if any difference in path length changes the observed measurement. The intensity of light can be reduced over distance as the photons interact with the air so attenuation of the signal may occur. The TILDAS system can extend to path lengths of $30m$ (Jimenez-Palacios, 1998) however the NDIR instrument does not produce the same coherent light beam and may be subject to additional signal attenuation. To ensure that the impact of path length is not causing any bias with the measurements the equipment was set up at three different separations. The instrument was set up at separations of 3.5m, 4.0m, 4.5m and 30 'gas puff' audit measurements were performed at each separation to ensure the most consistent measurement source possible. These path lengths are smaller than typically used on the road but the difference is representative of a decision made by the operator. Currently the setup locations are picked for reasons based on ease more than science however if the equipment is sensitive at these separations then operators will need to take more care when setting it up. If significant signal attenuation was observed steps will need to be taken to ensure that the path length is consistent.

The NO parts per million (ppm) measurements from this experiment are shown by distribution graphically in Figure 4.4 and the normal distribution coefficients are shown in Table 4.1. A green dashed curve along with a red dashed line indicate the expected distribution based on the calibration gas stated concentrations and instrument error quoted in ESP (2005). By looking at the results it seems that generally speaking the distribution of the measurements is the same regardless of separation. To better prove this a Kolmogorov-Smirnov test is applied to each of the individual separations and 1000 measurements generated randomly from the mean and standard deviation of the full set. The hypothesis that the two sets of data are derived from the same structure is rejected if $p < 0.05$. For 3.5m $p = 0.5997$, 4m $p = 0.5491$ and 4.5m $p = 0.3643$. A matrix of cross comparisons between the three data-sets is shown in Table 4.2. Natural variance is present in the measurements and this leads to the changes in observed distribution coefficients however it is statistically acceptable to treat the three different sets as coming from the same underlying distribution and as such it can be concluded that the difference in instrument separation or laser path length at the variations that are present between different on-road locations does not significantly influence the observed NO value measured by the RSD. This result is important as

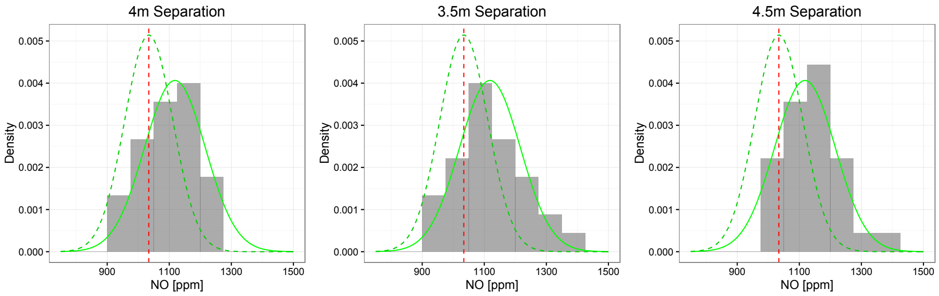

Figure 4.4: Distribution of NO over different separations.

Separation (m)	Mean (NO ppm)	Standard Deviation (NO ppm)
3.5	1115	112
4.0	1102	86
4.5	1137	94
Total	1118	98

Table 4.1: Table of normal distribution coefficients for different separations.

it means that the details of the site setup are less important than the measurements themselves.

4.2.4 Consistency and Accuracy of the Equipment

The data from the three separations were combined to form one large data set consisting of 90 data points that could be used to assess the consistency and accuracy of the instrument. An ideal normal distribution was defined based on the errors stated in the RSD man-

Separation (m)	3.5	4.0	4.5
3.5	1	0.594	0.808
4.0	0.594	1	0.594
4.5	0.808	0.594	1

Table 4.2: Matrix of K-S test p-values .

ual and the specified abundance of NO in the control gas canister and compared to the test data. The mean and standard distribution emissions were fitted to the NO measurements using R (R Core Team, 2015) and the *fitdistrplus* package (Delignette-Muller & Dutang, 2015). The fitting algorithm gives means and standard errors of 1118.2 ± 10.3 and 97.6 ± 7.3 for the mean and standard deviation respectively with an Anderson-Darling statistic of 0.51 which shows that there is a good chance that the data points were taken from a normal distribution (Anderson & Darling, 1952) hence the normal distribution is a good fit for the data as would be expected for a system without any internal bias and proving the hypothesis that the distribution of error introduced by measuring emissions using an RSD are normally distributed. It is therefore hypothesised that any change to this distribution in future measurements is a real physical factor relating to either the individual vehicle for repeated measurements or for the behaviour of the fleet when looking at a subset of different vehicles observed on real driving environments.

The results of the SDM validation experiment are displayed in Figure 4.4. A positive systematic offset is observed in the measured data compared to the gas bottle. The offset from the controlled gas bottle value may be a calibration issue for the RSD or may be an error introduced by the bottle itself as their relative abundances are subject to an error of 5%. The standard error for a normal distribution Δ is calculated for NO as $\Delta_{NO} = \sigma_{NO}/\sqrt{n}$ where σ_{NO} is the standard deviation and n is the number of NO measurements. In this case $\sigma_{NO} = 58ppm$ and $n = 90$ so $\Delta_{NO} = 6.1ppm$. The audit gas was therefore measured as $1118 \pm 6.1ppm$ by the RSD4600. The rated gas bottle value is $1034 \pm 52ppm$. These values are consistent with each other to greater than 98% (2σ) confidence. For NO the systematic error of the mean is $+84ppm$ and the 2σ, ($\approx 98\%$) confidence interval is $\pm 196ppm$. The correction falls within the error of the measurement however the percentage error is 18.9%, higher than the 15% quoted in the RSD instrument manual (ESP, 2005) but within the error of the abundances of the gas bottle. It is therefore concluded that the accuracy of the equipment is good.

4.2.5 Repeated Measurements of Vehicles' NO Emission

The previous section has shown that the results can be trusted to be within the limits of the instrument precision however it is important to show that the variation observed in repeated measurements of vehicles is because of the variation in emissions rather than observational error in the instrument. Repeated measurements of the same vehicles were undertaken both on-road and off-road. Off-road measurements were performed using a Euro 4 diesel passenger car (identified as Vehicle 6 in Table 4.4) as it had the highest number of measurements. It is hypothesised that the variance in measurements of NO emitted by Vehicle 6 will be greater than the variance in measurements observed by repeated measurements of the control gas NO concentration. If this hypothesis is shown to be false then the support for the RSD measurement to form part of the picture of vehicle emissions is significantly harmed as it will struggle to resolve anything more than the broadest of strokes.

Off Road Measurements of A Euro 4 Diesel Passenger Car

The Euro 4 diesel passenger car, a Honda Civic in this case, chosen for analysis is typical of fleet vehicles (See Figure 7.1) however it was chosen for this experiment as it had the greatest number of valid RSD measurements performed on it ($n = 171$). The equipment was set up as described in Figure 3.3. The vehicle was driven through the equipment multiple times and simulating different driving styles. The data presented focuses on the gear selection and driving styles. Measurements taken in gears 1 and 2 were selected with measurements taken in gear 3 discarded as too few valid measurements were collected.

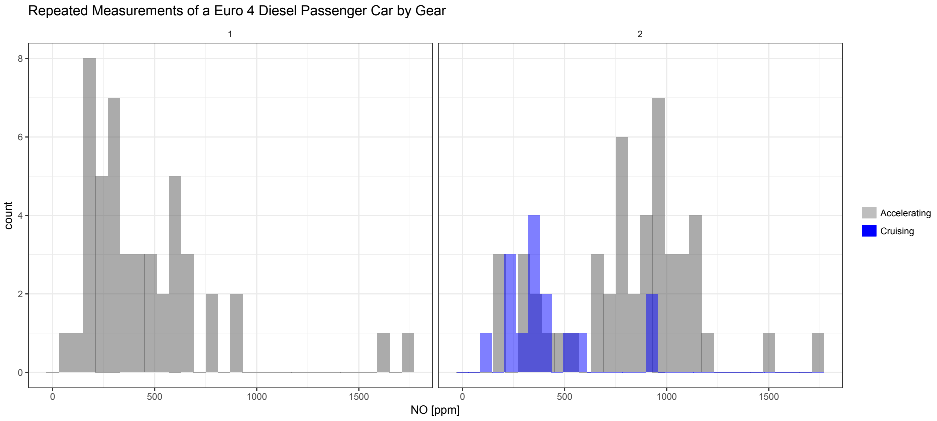

Figure 4.5: Distribution of NO over repeated measurements of a passenger car.

The vehicle was either accelerated through the equipment or cruised through the equipment at an engine speed comfortable to the driver so as to capture the natural variations found in real world driving. This method is more realistic than picking an engine speed or clock speed and using those measurements as guides as most experienced drivers concentrate primarily on the road rather than repeatedly scanning the instrumentation to achieve the target operation. The results for this experiment are shown in Figure 4.5. Acceleration measurements are shown in the grey histogram and overlaid with the constant speed measurements shown in blue. First and second gear are indicated in the panel titles. First gear cruising failed to provide any valid measurements most likely because of the low engine power requirement. In real driving environments it is rare to cruise in first gear so the lack of measurements here is not concerning. To better relate the results presented here the conclusions made in Section 4.2.4 the bin width is specified as one standard deviation of the equipment as observed in Section 4.2.4. The source of any variation beyond 2 bins is almost certainly due to the vehicle rather than the instrument.

There are three key points illustrated in this measurement. Firstly the RSD is sufficiently sensitive to see the natural variation in emission from the engine. This is evident because the spread of the measurements extends beyond 2 standard deviations and the distribution of measurements does not follow the normal distribution. Secondly the emission of a vehicle is shown to be dependent on the gear selected. Higher gears emit more than lower gears suggesting that the engine speed is less important than the power demand. Vehicles cruising produce less emission than those accelerating observed by the shift in peak emission in gear 2 to the right for cruising measurements. None of these results are particularly surprising however the key fact is that the RSD has sufficient instrument resolution to be able to identify these factors proving the initial hypothesis of this section. The conclusion is that when translated to whole fleet measurements it is reasonable to say that any distribution observed is as a result of the fleet measured rather than variation caused by the instrument itself. This conclusion forms the basis for the work done in Chapter 5 and beyond.

On Road Measurements of Busses

Diesel busses are generally understood to emit a significant amount of NO and understanding how these emissions are distributed is important to identifying very highly emitting vehicles that may have faulty systems (Tate, 2016, 2013a,b). Repeated measurements of highly emitting vehicles have been performed previously (Bishop *et al.*, 2009) and the previous section shows that the RSD is capable of resolving the variation in repeated measurements from fleet vehicles. This section will attempt to show that the RSD is capable of resolving the distribution of emissions from high emitting vehicles as well as more typical fleet vehicles. The only vehicles captured regularly enough on the road to be useful in this analysis were busses. The site Eyre Street from the Sheffield data set (Section 3.4.1) was the site with the most

Bus	Euro Fuel	Number of Measurements
1	Euro 4 Diesel	49
2	Euro 4 Diesel	48
3	Euro 4 Diesel	30

Table 4.3: Table of busses used for repeated measurement analysis.

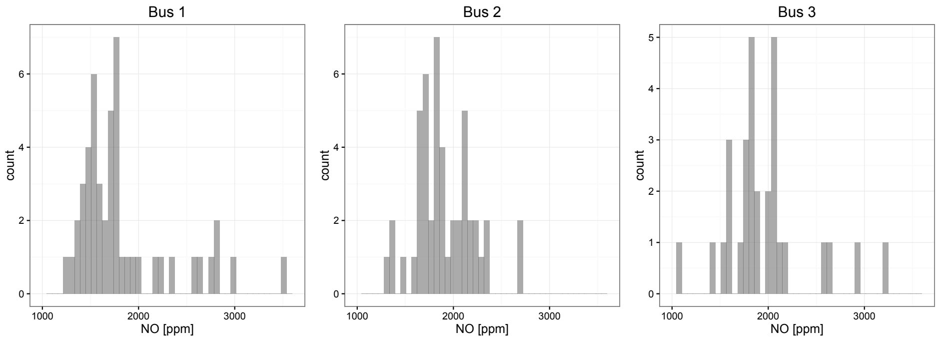

Figure 4.6: Distribution of NO over repeated measurements of three busses in Sheffield.

busses measured. The three busses with the most total measurements over the two observation days were selected for further analysis. The vehicle descriptions are presented in Table 4.3 and their respective emissions are presented in Figure 4.6.

As with the repeated measurements of vehicles performed off road the histogram shows one standard deviation of the measurement error of the remote sensing device. Less metadata is available relating to the operating conditions of these busses however they were driven on the same route and the measurement site was located right after a junction so they represent the most consistent set of results possible with on-road deployment. Again the RSD shows that it is capable of measuring variance in the NO emissions beyond and that it is beyond that of the instrument. If the instrument was unable to resolve the variance of the busses' real driving emissions then any steps towards modelling it would result in simply modelling the instrument. Showing that the distribution of emissions observed by the instrument is not significantly influenced by the instrument itself is important going forward modelling the vehicle emissions. The measurements taken by the RSD have been shown to be accurate enough to not lose information about the distribution of vehicle emissions and therefore any steps to model these data are valid for both highly emitting vehicles and vehicles more representative of the passenger vehicle fleet. It is also interesting to note that in real driving environments there is a significant variation not only between vehicles but for the same vehicle. This has implications for those who are modelling individual vehicles and extrapolating this result to the fleet as a whole.

4.2.6 Validation of NO Measurements

An opportunity presented it's self during the course of the research programme to cross validate the RSD4600 with the prototypical FEAT system developed at the University of Denver. Whilst an attempt has been made by Bishop *et al.* (2009) to cross validate the two instruments the study was performed on heavy commercial vehicles travelling at slow speeds after a weigh bridge in the USA. The results of this comparison showed that there was an underestimate of $27\% - 30\%$ in the NO measured by the RSD4600 ($R^2 > 0.78$) that they used compared to the FEAT system that was taking simultaneous and colocated measurements. Repeating this experiment with a wider range of vehicles and a different RSD4600 would be able to show if this was an underestimate consistent with multiple RSD4600 units or an isolated systematic error.

The Denver Fuel Efficiency Automobile Test (FEAT) instrument uses a Non Dispersive Infrared (NDIR) laser system and a Dispersive Ultraviolet laser system. The systems consist of a dual element light source (silicon carbide gas drier igniter and a xenon arc lamp) and a separate detector unit with four non-dispersive infrared detectors that provide an IR reference (3.9 μm) and measurements of carbon dioxide (CO_2, 4.3 μm) as well as channels for CO and HC measurements not used in this paper. The detector unit is connected by fibreoptic cable to two dispersive ultraviolet spectrometers that measure NO, sulphur dioxide (SO_2) and Ammonia (NH_3) between 200nm and 226nm, and NO_2 between 430nm and 447 nm (Carslaw *et al.*, 2015). In this instance the two units are powered by two petrol generators located approximately 5m from their respective instrument on each side of the road increasing the risk of contamination of the emissions measurements by transient sources of pollution. The RSD4600 uses an NDIR system operating at the same frequency windows as the FEAT system without the facility to measure NO_2, SO_2 or NH_3. Unlike the FEAT system a corner cube mirror (CCM) is deployed for the RSD4600 system such that the sensing beam is reflected back and the path length is doubled, hence increasing the number of interactions with beam photons and signal to noise ratio, potentially improving the accuracy of the measurement. Having the source module and the detector module on the same side of the road with the same power supply and connected directly to the control unit means that the system as a whole is more stable. More accurate measurements of the source strength can be achieved. Variables related to atmospheric and ambient conditions amongst others can be better accounted for and any related systematic errors can be removed. Contamination of the signal by transient sources is mitigated through the use of a dedicated van and a large portable battery power supply. The battery packs mounted in the van guarantee a consistent power supply throughout long periods of measurement.

Along with the opportunity to test the RSD4600 the FEAT system has capabilities that the RSD4600 does not and the opportunity to use the two side by side might extend the capabilities of the RSD4600 in estimating total NO_X, a significant goal of this thesis. The FEAT system is able to measure Nitrogen Dioxide (NO_2) directly and hence calculate the fraction of total NO_X or the f_{NO2} value required to upscale $NO : CO_2$ to total NO_X.Being able to take a direct measurement of f_{NO2} is one of the main limitations of the RSD4600 and if the FEAT measurement of NO_2 could be related to the RSD4600 measurement of NO then previously measured f_{NO_2} values could be used to give more valid estimates of the total NO_X measured by the RSD4600 can be made. Before taking this step it is important to show that the two measurements of NO are consistent otherwise any NO_2 measurements used for estimating NO_X would not be valid when used alongside the RSD4600 $NO : CO_2$ measurements.

The instrumentation was set up as described in Section 3.3.3 and shown in Figure 3.3. A selection of vehicles of different Euro Class, fuel type and vehicle class were repeatedly driven through the equipment. A summary of these vehicles is presented in Table 4.4. Simultaneous measurements of the NO were measured and the FEAT system also measured f_{NO2}. The clocks were synchronised and the measurements where a valid NO measurement from both sets of equipment were used to form a correlation.

The measurements from the two instruments should have a linear relationship if they are well correlated. A linear model with the form $y = c_1x + c_2$ is fitted to the data with the coefficients $c_1 = 0.93$ and $c_2 = 0.0$ being derived from the fit. The adjusted R^2 value is 0.85. These values are consistent with the two instruments being well correlated. Some natural variation is expected and observed. There is no evidence of the systematic decrease observed in Bishop *et al.* (2009) and the R^2 value suggests a better correlation than was previously observed. The results are displayed in Panel a of Figure 4.7 in blue with a 95% confidence interval shown in grey. The 1:1 relationship is identified with a red dashed line. The RSD4600 $NO_X : CO_2$ is calculated using Equation **??**. The f_{NO2} values for each vehicle are estimated using two methodologies. The middle panel uses the values

Vehicle	Fuel	Euro Class	Type	Valid Measurements
1	Gas / Bi Fuel	3	LCV (Heavy Van)	83
2	Petrol	4	Car	39
3	Diesel	5	Car	22
4	Diesel	4	Car	45
5	Diesel	4	LCV (Car Derived)	18
6	Diesel	4	Car	171
7	Gas / Bi Fuel	5	LCV (Medium Van)	30
8	Diesel	5	Car	23
9	Diesel	4	LCV (Car Derived)	18
10	Diesel	2	LCV (Heavy Van)	21
11	Diesel	4	LCV (Heavy Van)	26
12	Diesel	4	LCV (Heavy Van)	32
13	Diesel	5	HCV (Rigid 2 Axel)	17
14	Diesel	4	Bus (Single Deck)	12
15	Diesel	3	LCV (Medium Van)	109
16	Diesel	5	Car	25
17	Petrol	4	Car	7
18	Diesel	4	Bus (Single Deck)	19
Total	-	-	-	717

Table 4.4: Description of individual vehicles used in GPS and cross-instrument analysis.

derived by Grice *et al.* (2009) and the right panel uses the values measured by Carslaw & Rhys-Tyler (2013) using the FEAT system but at an earlier date than and in no way related to the measurements taken with the RSD4600. In each case the vehicle type, fuel and euro class values displayed in Table 4.4 are used to estimate each vehicles' f_{NO2} individually. The f_{NO2} values are shown in Table 4.5.

4.3 Derivation and Validation of New Total NO_X Calculation for RSD4600

As shown in Section 4.2.6 the RSD4600 and FEAT system give largely consistent measurements of NO and hence the f_{NO2} measurements previously taken by the FEAT system can be used as a basis for calculating total NO_X using the RSD4600. The method remains to be validated however and this section will show how the relationships are derived and will attempt to test and validate the new methodology.

4.3.1 Methodology

Equation 3.3 shows how the f_{NO2} and the NO measurements relate to the total NO_X. Previous attempts to relate these measurements have been relatively successful but relied on the *Netcen* model to predict the f_{NO2} based on the interaction of NO, NO_2 and O_3 (Abbott & Stedman, 2005; Grice *et al.*, 2009) a method commonly applied in such calculations. The RSD method allows us to directly tie the f_{NO2} measurements to the total NO_X. Grice *et al.* (2009) attempted to use this methodology and the factors derived in this work have been used in multiple UK based projects.

As part of a project performed in London f_{NO2} measurements were made using the FEAT system prior to the colocated measurements being taken (Carslaw & Rhys-Tyler, 2013). These factors are aggregated based on vehicle type, fuel type and euro class. A summary is

Vehicle Type	Euro 1	Euro 2	Euro 3	Euro 4	Euro 5
Petrol Car FEAT	0.02	0.02	0.03	0.04	0.08
Diesel Car FEAT	0.14	0.08	0.16	0.28	0.20
Light Commercial FEAT	0.11	0.09	0.11	0.26	0.25
Petrol Car Old	0.04	0.04	0.03	0.03	0.33
Diesel Car Old	0.11	0.11	0.30	0.55	0.55
Light Commercial Old	0.11	0.11	0.30	0.55	0.55

Table 4.5: Fractional NO_2 values used to calculate total NO_X measured using the FEAT instrument previously with previous coefficients in parentheses.

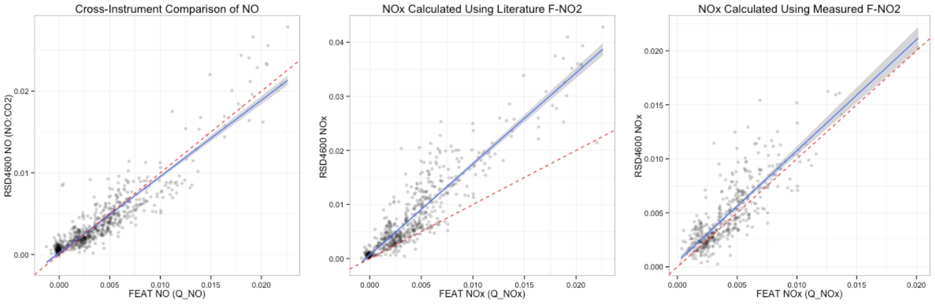

Figure 4.7: Correlation of instruments for NO and total NO_X.

provided in Table 4.5 along with the previously used coefficient presented in parentheses. There are some notable differences between the coefficients so it would be expected that the two methodologies would produce different results. The total NO_X is calculated using both sets of coefficients and the NO measurement performed by the RSD4600 and compared to the total NO_X measured for each vehicle using the FEAT system using currently measured f_{NO2} coefficients.

4.3.2 Results

The results for this analysis are presented in Figure 4.7. In the centre panel the old method for calculating total NO_X using RSD4600

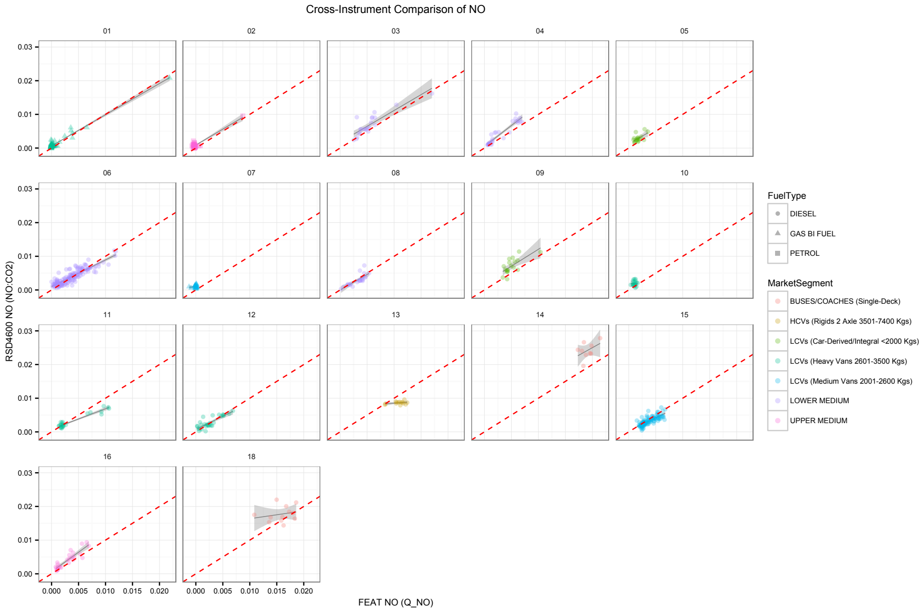

Figure 4.8: Individual vehicle comparison of both instruments' measurements of $NO : CO_2$ ratios

compared to measured NO_X is shown. In the right panel the new method for calculating total NO_X using on-road measurements of f_{NO2} using the FEAT system and RSD4600 NO measurements is shown. Both methods show a reasonable correlation with the measured total NO_X however the method using measured values gives a much better one-to-one correlation.

It has been shown that the NO measurements between instruments produce a strong linear relationship and that the method of remote sensing f_{NO2} to calculate total NO_X produces a better correlation than the previously used method. To remove any uncertainty when applying this model each vehicle was examined individually to show any biases that might be present due to the different vehicle and fuel types used by fleet vehicles. Vehicles are described in Table 4.5 and the results are presented in Figures 4.8 and 4.9 along with a map of the ideal one to one relationship displayed as a dashed red line.

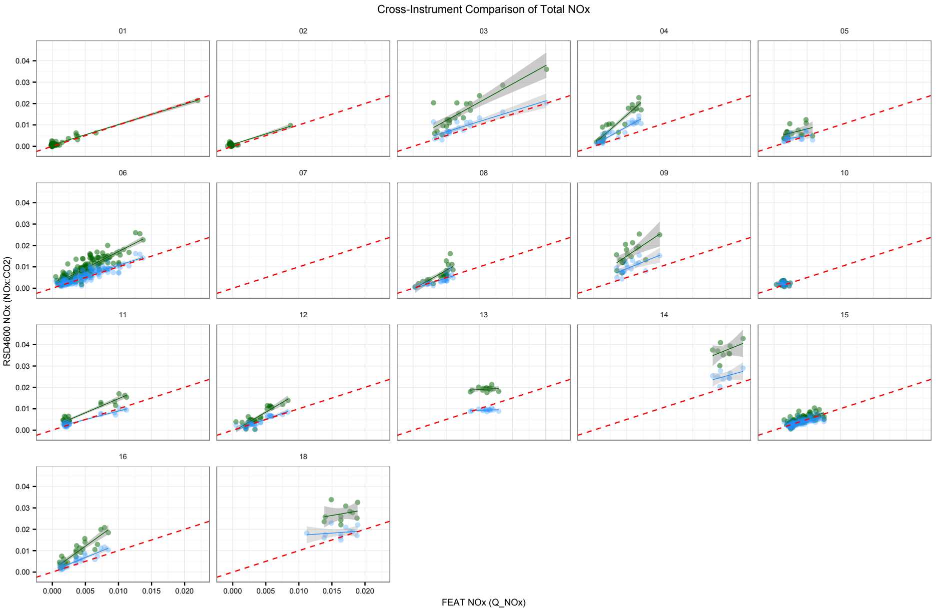

Figure 4.9: Individual vehicle comparison of both methods for calculating total NO_X : CO_2 ratios with the old method shown in green and the new method shown in blue

Metadata relating to the vehicles' fuel type and class are presented only in Figure 4.8. Linear models are fitted to each vehicles plot with a 95% confidence interval and this is indicated by the shaded area.

The previously observed relationship between the RSD4600 and FEAT NO ratio emissions are confirmed and shown to be unbiased to any particular type of vehicle or fuel type further strengthening the case that the instrument correlation is consistent across instruments with different specifications. For the majority of vehicles the new (blue) method for calculating total NO_X is an improvement over the old (green) method. In some cases a small systematic overestimation by the RSD4600 is observed such as is present in vehicle 4, vehicle 9 and vehicle 14 however in comparison to the old method these still represent an improvement. Vehicle 1 and vehicle 2 do not have fractional NO_2 values and as such cannot be measured however in these cases the old method remains very effective at predicting the total NO_X. The case of vehicle 13, a rigid axel heavy commercial vehicle, which is of special interest to many groups may prove very useful moving forward.

For the majority of vehicles the RSD4600 $NO : CO_2$ measurements alongside the NO_{X-FEAT} method using measured f_{NO2} values for calculating total NO_X correlates strongly with the side by side FEAT measurements. A χ^2 goodness of fit test was applied to both the methods giving results of $\chi^2_{model} = 3.83$ and $\chi^2_{FEAT} = 0.371$ suggesting that the FEAT method of deriving total NO_X is superior to the previous methodology. By looking at the correlation plots it is clear that for most cases the FEAT method is an improvement over the $NO_{X-model}$ method using modelled f_{NO2} values and at worst an equivalent result. This is especially evident in higher emitting vehicles. In some cases a small systematic overestimation by the RSD4600 is observed such as is present in vehicle 4, vehicle 9 and vehicle 14 however in comparison to the old method these still represent an improvement. Vehicle 1, a modified petrol LCV, and vehicle 2 do not have fractional NO_2 values available and as such cannot be estimated using the FEAT method however in these cases the $NO_{X-model}$ method remains very effective at predicting the total NO_X and the results are included to reflect this. The cases of vehicles 13 and 4, rigid axle heavy commercial vehicles, show that the old methodology for estimating its NO_X emission is an underestimate by a factor of 2. The influence of commercial vehicles in total NO_X emission is of special interest to many groups and this result may prove very useful when calculating total NO_X emission inventories moving forward.

4.4 Conclusions

The experiments detailed in this Chapter set out to validate the measurements performed by Remote Sensing Device based studies using the RSD4600. The speed and acceleration measurements were validated using a state of the art GPS system with the RSD4600 performing strongly. Very good correlation was observed for the speed measurement and good correlation was observed with the acceleration measurement. As the speed and acceleration module performed well studies using the speed and this element of the RSD4600 to calculate vehicle specific power as a diagnostic tool are validated. The results are limited to vehicles operating in the lowest three gears however for urban driving these results represent the majority of operating conditions. Additional measurements of vehicles travelling in fourth gear were not available due to space restrictions at the testing venue. These measurements would prove useful for a full range of vehicle operating conditions and would form an interesting further research project however the range of VSP that can be calculated from this set gives a representative range of VSP observed in real driving environments.

The performance of the RSD4600 has been rigorously tested under controlled conditions and has also had its results validated. An unaccounted for bias of $+84ppm$ has been found in the calibration or results of the controlled gas experiment however all results fall within the tolerances of the equipment and the materials used. At the current time because the source of the bias cannot be isolated it is not treated as part of the systematic error of the instrument but as an error relating to the materials. It is hypothesised that the source of this error is due to the uncertainty in control gas abundances. Any systematic error caused by uncertainties the control gas abundances would be systematically carried through the distribution analysis rather than adding to the measurement error. Repeating the experiment with a different gas bottle would show whether or not the error is due to uncertainties in the control gas abundances or a calibration problem with the instrument.

The NO measurements made by the RSD4600 of vehicles passing through the equipment have been compared with the FEAT system on loan from the University of Denver. Side by side comparisons of a wide range of vehicles have been made with the RSD4600 matching almost perfectly. The strong correlation with no statistically significant offset suggests that the systematic error observed previously is probably due to the gas bottle as different calibration gas was used between instruments. This result is an improvement over previously reported results which suggested that the RSD4600 consistently underestimated the $NO : CO_2$ ratio present in the exhaust plume. It is unclear what the source of this error was in previous measurements however for the data collected throughout the rest of this thesis it is assumed that no additional correction is required at the instrument level. In addition to this finding the consistency between instruments suggests that the unaccounted for bias previously identified is less likely to be at the instrument level as any bias would be observed in the intra-instrument measurements. Given that previous studies have identified the bias as negative it seems even less likely that the source of the positive bias observed in these results is the instrumentation. On the balance of probabilities it is more likely that the source of the observed bias is caused by an error in the relative abundances of gasses in the control gas cylinder.

The relationship between the instruments' NO emission and total NO_X validation shows that the instruments are consistent across a range of measurement conditions. The results have shown no bias towards or against any particular vehicle type or fuel class with consistently well correlated results regardless of vehicle tested. The most significant implication is that fractional NO_2 (f_{NO2}) values measured in previous studies by the FEAT system, which the RSD4600 cannot measure, can be used to study the total NO_X emitted by vehicles in RSD4600 studies without the logistical trouble and cost associated with transporting the FEAT system to the UK in future. The consistency across the range of measurements means that a new method for calculating total NO_X emissions using remotely sensed fractional NO_2 could be created and validated against an instrument that could measure it directly. Validation of the f_{NO2} measurements using this method has also been shown to be superior to methods using roadside models previously used. The method has shown noticeable improvements in the correlation with heavier duty vehicles such as LCVs and HCVs as well as with busses. These are high emitting vehicles that are of particular interest to policy makers. With the interest in contribution by vehicles other than passenger cars to the pollution inventory this study along with previous work done only using the FEAT system to provide these factors give a useful tool kit moving forward. This methodology is limited as no Euro 6 measurements were made using the FEAT instrument so no remotely sensed f_{NO2} observations are available however the success of the methodology where f_{NO2} measurements are available means that additional sources of measurements may be used with some confidence. When required throughout the rest of the thesis values are sourced from Heijne *et al.* (2016).

It is concluded that in the future the RSD4600 can be used in urban settings to accurately describe the total NO_X emissions from a wide range of vehicles not limited to passenger cars but including light commercial vehicles, heavy commercial vehicles and busses with a high degree of confidence. The new total NO_X estimations can also be applied to previous studies improving the size of the database of total NO_X emissions measurements available to researchers in the future. These conclusions continue to bolster support for the notion that use of remote sensing in urban driving vehicle emission decision making is a valid and useful part of a holistic process aimed at understanding the source of urban air pollution and thereby allowing policy makers to make better informed decisions relating to the reduction of total NO_X concentration in real driving environments. In the context of this thesis the conclusions drawn from this chapter are important. They mean that the emissions observations presented throughout the rest of the thesis can be used to estimate a total NO_X emission factor more accurately than previously possible.

Chapter 5

Extreme Value Mathematics and Modelling Vehicle Emissions

5.1 Introduction

A solid mathematical framework for the description of any phenomena is a critical first step to developing a numerical model. Models of vehicle emissions that use a mean and standard deviation as the input parameters are not sufficient. Neither the distribution of vehicle emission ratios or emission factors are symmetrical and therefore basic statistics relating to the mean and standard deviation are too simplistic to capture the nuances of the fleet behaviour. Any statement such as the average NO_X emission of Euro 4 vehicles has increased from Euro 3 is useful only if no further analysis is available. This model is of little use when trying to describe the data in depth as any summations or aggregations performed will fail to account for the asymmetry in the real world emission factors. Extreme value distributions allow asymmetrical data such as those observed in vehicle emissions data to be described in a robust mathematical way. The Aberdeen Euro 5 diesel passenger car fleet is fitted to the normal distribution in four different ways to show that this function is not appropriate for this distribution of emissions factors in Figure 5.1. A Probability Density Function (PDF) plot shows the probability of a value taking a given value with the observed data binned as a reference. As with the PDF the Cumulative Density Function (CDF) plot shows the same information but the probabilities are summed up cumulatively with points indicating the observed data. A Quantile-Quantile (Q-Q) plot shows the theoretical and the observed quantiles of the data with data matching the model perfectly when a straight line is observed. The Percentage-Percentage (P-P) plot compares the theoretical and observed CDFs and as with the Q-Q plot when the theoretical and observed data match the result is a straight line. The plots in Figure 5.1 were plotted with the R (R Core Team, 2015) package *fitdistrplus* (Delignette-Muller & Dutang, 2015). The deviation from the theoretical model present in both the Q-Q and P-P plots as well as a failure to match the theoretical CDF and PDF functions predicted when the normal distribution is applied to the data in Figure 5.1 show that the normal distribution is not appropriate for modelling whole-fleet vehicle NO emission ratios and also the NO_X emission factors as they are estimated using this raw measurement.

A series of events with rare but high value events can be characterised by extreme value distributions. Various forms of extreme value distribution have been applied to many real world scenarios where the distribution of the scale of events does not follow a normal distribution. The use of the extreme value distribution extends from finance (Bensalah *et al.*, 2000; Poon *et al.*, 2004) to hydrological data (Martins *et al.*, 2000). Generalised extreme values (GEV's) have been used extensively in choice modelling with Daniel McFadden receiving the Nobel Prize for Economic Science in 2000 for work in this field. The work has been extended throughout choice modelling and has been used in a number of different transport related scenarios (Bierlaire *et al.*, 2008; Daly & Bierlaire, 2006; Ma & Fukuda, 2015).

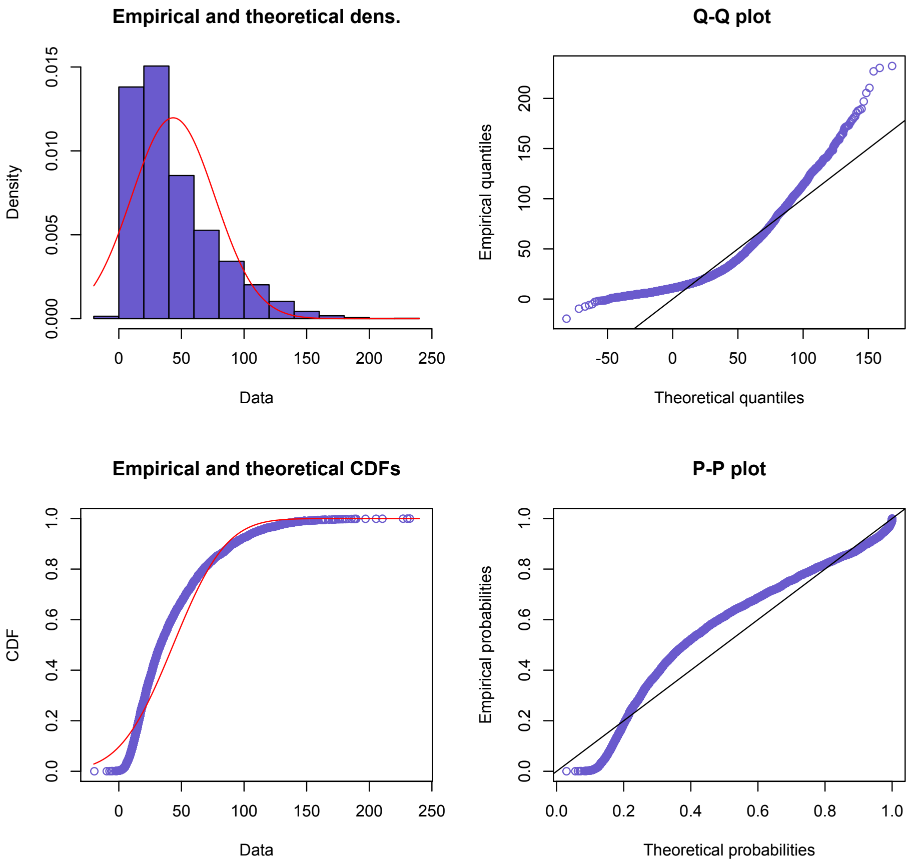

Figure 5.1: Aberdeen Euro 5 diesel passenger fitted to a normal distribution

There are three different types of extreme value distribution. These are called Weibull, Frechet and Gumbel. The Weibull distribution (Equation 5.1 for $x < 0$) was first identified by Fréchet (1928) and first used by Rosin (1933) for analysis of the sizes of coal particles. The Weibull distribution is defined by the shape parameter and the scale parameter and is often used in materials science to analyse the failure rate. The shape parameter (k) has three potential values. Where $k < 1$ a material has a high 'infant mortality' and failure rate decreases over time. Where $k = 1$ the rate of failure is independent of time suggesting that some external random events are responsible for the failure. Where $k > 1$ the failure rate increases with time indicative of ageing processes being responsible for the failures. The Frechet distribution (Equation 5.2, also known as the inverse Weibull distribution is defined in terms of a shape parameter a location parameter and a scale parameter. It is often used in hydrology to predict maximum river discharges and maximum daily rain fall (Coles *et al.*, 2001).

$$P_{Weibull} = \frac{k}{\lambda}(\frac{x}{\lambda})^{k-1}e^{-(x/\lambda)^k} \tag{5.1}$$

$$P_{Frechet} = \frac{\alpha}{s}(\frac{x-m}{s})^{-(1+\alpha)}e^{-(\frac{x-m}{s})^{-\alpha}} \tag{5.2}$$

The Gumbel distribution (Gumbel, 1935, 1941) , also known as the log-Wiebull distribution. The Gumbel distribution is defined in terms of its scale and position, with the probability density function described in Equation 5.3 where $z = \frac{x-a}{b}$, and has been used to model and predict rainfall and river discharge. In mathematics the Gumbel distribution is associated with the number of terms in the partition of an integer (Erdos & Lehner, 1941) and the size between prime gaps (Kourbatov, 2013). The Gumbel distribution is especially useful for describing vehicle emissions as it can relate a sample to both a peak value analogous to a mode through the location parameter and the impact of the higher emitting vehicles in the scale parameter. The Gamma distribution is not of the same family as the EVD functions described previously but has also been used in rainfall predictions (Aksoy, 2000) and actuarial science (Boland, 2007). A comparison to the Gamma distribution that has also been used to describe the vehicle emissions (Zhang *et al.*, 1994) distribution is shown in Figure 5.2. The Gumbel distribution has a technical advantage over the gamma distribution, Equation 5.4 where the shape parameter is α and the rate parameter is β with $\alpha, \beta, > 0$ and $\Gamma(\alpha)$ is the gamma function of α, in that it allows for the negative values measured by the RSD, statistically important for avoiding bias, to be counted without having to apply mathematical transformations the data set. These negative values are valid as the instrument error (Section 4.2) means that if the absolute value is low then some values will be negative. Removing these data points as invalid from the set would introduce a positive bias to any aggregated calculations performed. Conceptually the Gumbel distribution provides a clearer picture as it's parameters are more closely related to physical parameters than the Gamma distribution. The Gumbel location parameter a is equivalent to the mode of the distribution and has the same units of measurement. Figure 5.2 shows a selection of probability density distribution functions for both Gumbel and gamma distributions.

$$P_{Gumbel} = \frac{1}{b}(e^{z+e^{-z}}) \tag{5.3}$$

$$P_{gamma} = \frac{\beta^{\alpha}}{\Gamma(\alpha)} x^{(\alpha-1)} e^{-\beta x} \tag{5.4}$$

Attempts have been made to quantify the roadside concentrations of pollutants in terms of the extreme value distribution (Mdyusof *et al.*, 2011; Sharma *et al.*, 1999). Attempts have also been made to quantify vehicle emissions in terms of the Gamma distribution (Zhang *et al.*, 1994). Vehicles that exist in the high emitting range may contribute a significant amount of the total fleet emission (Bishop *et al.*, 2016). The results presented in Zhang *et al.* (1994) have rarely been cited and

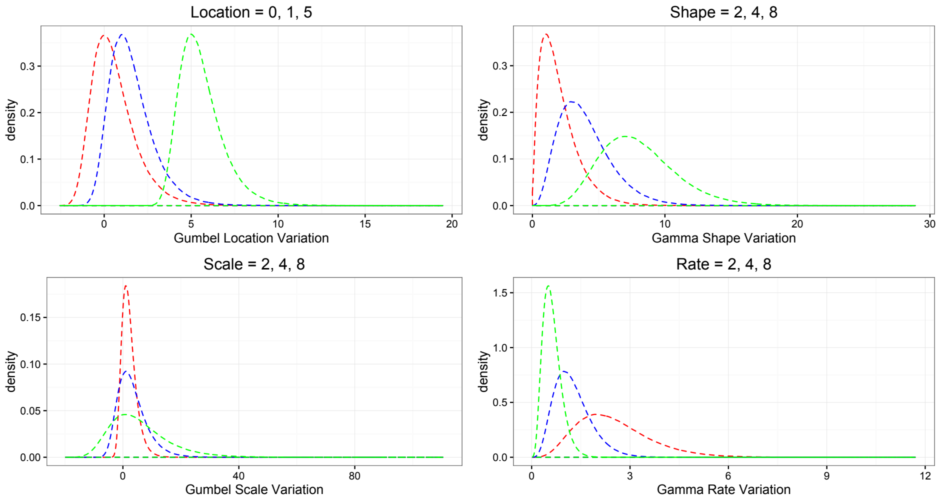

Figure 5.2: Variations in parameters for Gumbel and Gamma distribution

the implications of the use of an asymmetrical distribution to whole fleet vehicle emissions factors is currently unclear. Extreme value distribution analysis, using the Gumbel, Weibull or Frechet distribution functions, has not been applied to vehicle emissions factors. The use of this method to provide quantitative descriptions of vehicle emission data is the focus of this section. Section 5.4 then describes the development of a model using RSD data based on this work.

A number of tests will be performed that demonstrate the capability of the extreme value distribution to present quantitative statements about the differences in NO_X emissions by different subsections of the vehicle fleet. The whole range of datasets described in Section 3.4 are used. The Zurich data is used when all cold starts must be removed and vehicles should be running optimally. The Leeds data set is used to contrast the Zurich data set. The Sheffield and Cambridge data sets are used to compare and contrast the VSP and different sites as they have significantly different gradients across the range of observation locations. The Aberdeen data set is used when Euro 6 vehicles are to be analysed as it is the only one of the data sets that has a significant number of these vehicles present. It will show that the emission distribution of the vehicle fleet are of two types with the majority of vehicles following a Gumbel distribution (described as 'on-model') with an additional small fraction of vehicles exhibiting 'off-model' behaviour by very extreme emission factors that would not have been predicted by the model. The reasons for this are explored and analysed. A basic predictive model is described. The model shows how this descriptive process can be transformed into a predictive tool giving policy makers an idea of how various intervention strategies can be used to impact the total NO_X emitted by the whole fleet. A number of possible limitations of this model are tested and explored.

5.2 Application of Extreme Value Distribution to Vehicle Emission Data

5.2.1 Initial Fits

Five candidate distributions were selected to describe vehicle emissions: the Gumbel, Gamma, Weibull, log-normal and normal distributions. These were selected as they provided a range of different shapes of distribution function. The dataset made available by a team working from Zurich (Section 3.4.5) was used as a testing platform. These measurements were chosen as they represented the data with the least driver and vehicle dependant variables. The data was taken after a long period of uphill driving with vehicles fully warmed up eliminating a lot of the potential influencing factors described in Section 5.3 and serving as the closest thing possible to a control group. This site selection is also important as it means that the vehicles are most likely to be operating at their optimum temperatures and any particle filters more likely to have been regenerated before measurements were taken.

For initial comparison the $NO : CO_2$ ratio for Euro 4 diesel vehicles were chosen and the following distributions were fitted to them using the R (R Core Team, 2015) package *fitdistrplus* (Delignette-Muller & Dutang, 2015): normal, log-normal, gamma, Weibull and Gumbel. An offset of +130 is applied to each fitting routine to ensure that all negative values are removed as is required by the log normal and gamma distributions to ensure that all fits can be compared like for like. The resultant Q-Q comparison plots are displayed in Figure 5.3. The gamma distribution has a shape parameter of 3.43 ± 0.06 and a rate parameter of $4.8 \times 10^{-3} \pm 0.9 \times 10^{-5}$ the Gumbel distribution has a position parameter of 408 ± 4 and a scale factor of 293 ± 4 where the pm error is given by the standard error returned by the fitting procedure. Interpreting Q-Q plots is best performed by inspection with results closest to the equivalency line chosen. Based on this criteria both the gamma distribution and Gumbel distribution are good descriptions of the Euro 5 and Euro 4 diesel vehicle population and use of the Gumbel distribution can be used to describe vehicle emissions. Figure 5.3 also confirms the statement in the first paragraph of this section by demonstrating that the normal distribution, defined in terms of the mean and standard deviation, is not appropriate for an in-depth description of the distribution of the data. The Euro 6 diesel vehicles do not behave in the same way as the Euro 5 and 4 vehicles. This phenomena is further explored in Section 5.4.2 and beyond.

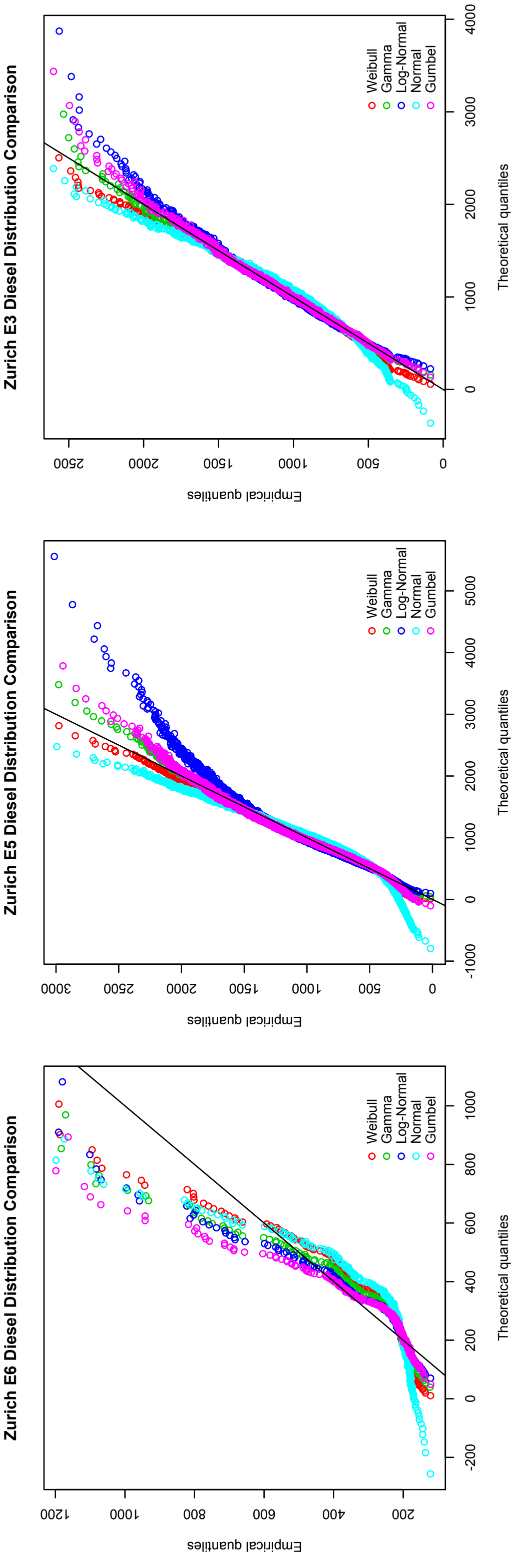

Figure 5.3: Zurich E6, E5 and E4 Diesel $NO : CO_2$ Q-Q Comparison plot

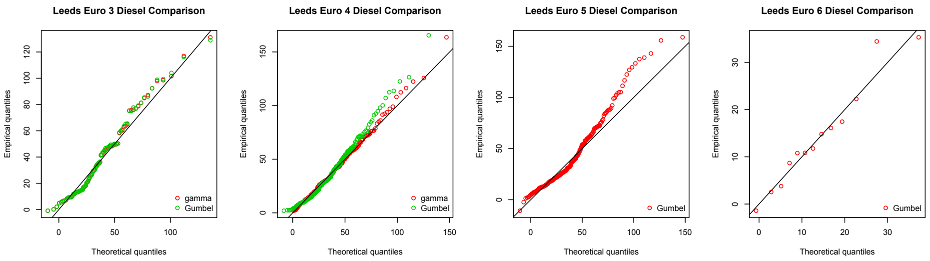

Figure 5.4: Leeds E6, E5, E4 and E3 Diesel $NO : CO_2$ Q-Q Comparison plots

To further validate the use of the Gumbel distribution in modelling vehicle emissions in urban environments data collected from Leeds (Section 3.4.3) for comparison in Figure 5.4. This data set was chosen as it represented a different real driving environment than the Zurich set. If the assertion that vehicle emissions can be modelled using the Gumbel distribution is to be upheld then the model must work in different circumstances. Where possible a gamma distribution is also presented for comparison however the fitting algorithm was unable to produce a gamma distribution for some examples as the gamma distribution requires all observations to be positive. The inability for the gamma distribution to fit the observed data without transformation in a lot of cases, the evidence that the gamma distribution is not following the Gumbel distribution in urban environments and the success of the Gumbel distribution in describing the vehicle emissions by it's self has meant that the Gumbel distribution is selected as the primary distribution to be used. In Section 7.4 the gamma distribution is presented alongside the Gumbel and shown to be inferior when describing the emission factors of taxis. Neither distribution is perfect however and for predictive purposes additional steps must be taken to ensure a successful result when using the gamma distribution. The observation of a small number of off-model vehicles is noted here. Their deviation from the Gumbel distribution will be addressed in Section 5.4.2.

In Figure 5.3 and Figure 5.4 it has been demonstrated that the distribution of the $NO : CO_2$ ratio emitted by diesel cars featuring a range of different emission abatement technology and encompassing three and four different iterations of emission reduction regulations as measured by the RSD4600 in both the UK and in Switzerland can be successfully modelled using the Gumbel distribution. Furthermore it has become very clear that the vehicle fleet naturally splits into two distinct sections. Those that do follow the Gumbel distribution very well and those that do not. It has been asserted that these vehicles behave differently and are extreme emitters because of faults and a wide range of other factors that are influenced either by age or sub-optimal operation of the emissions limitation systems for example cold starts. The remote sensing device deployed in survey mode is not the correct tool to diagnose these problems however this methodology is considered useful in identifying vehicles with abnormal emissions behaviour and potentially faulty systems.

Diesel vehicles are of more interest to policy makers as they emit more total NO_X than their petrol powered counterparts. It remains important to understand how petrol vehicles behave as a population if any serious attempt is to be made at modelling the passenger car fleet as a whole. The $NO : CO_2$ ratio of petrol fleet present in the Zurich data set was also analysed in the same way as the diesel fleet to attempt to understand how it behaves as a whole. The resulting Q-Q plots are presented in Figure 5.5.

The results of this analysis suggest that the fleet of petrol vehicles do not behave in the same way statistically speaking as the diesel fleet when observed at the Zurich site. A large number of vehicles are behaving as off-model extreme emitters when framed by the Gumbel distribution compared to the diesel fleet. This result is surprising as petrol vehicles and their three way catalysts are generally regarded as the more effective method of pollution removal and a better correlation

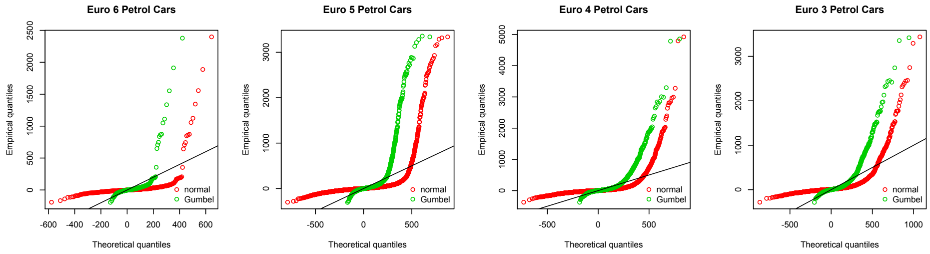

Figure 5.5: Zurich E6, E5, E4 and E3 Petrol Q-Q Comparison plot

with the distribution might be expected. It is worth noting however that the vast majority of the most modern Euro 6 vehicles do follow the extreme value distribution and the loss of correlation is caused only by the most extremely high emitting members of the population. There are a number of potential reasons for this deviation. Firstly the diesel vehicles running under different conditions to those experienced by urban vehicles may be closer to their optimum operating window and hence the emissions control systems are functioning better. The fact that occasional petrol vehicles emit very high levels of emissions suggests that this may be due to a fault, modification or other ageing process in the emission control system or the vehicle experiencing suboptimal operating conditions.

The petrol vehicles from the Aberdeen dataset were given a total NO_X emission factor based on the fractional NO_2 values measured by Carslaw & Rhys-Tyler (2013) with their CO_2 values uplifted in line with the most up to date uplift values available (Hagman & Amudsen, 2013; Tietge *et al.*, 2015; Weber *et al.*, 2015). Equation 3.5 shows the calculation of total NO_X emission factor and is the general case for any RSD measurement where ϕ is the uplift factor derived from the previously cited sources. The distribution of their emissions was analysed in the same way as previously demonstrated in this section. To better understand the role of the super-high emitting vehicles present in the fleet they were cut from the data-set with the resultant set pre-

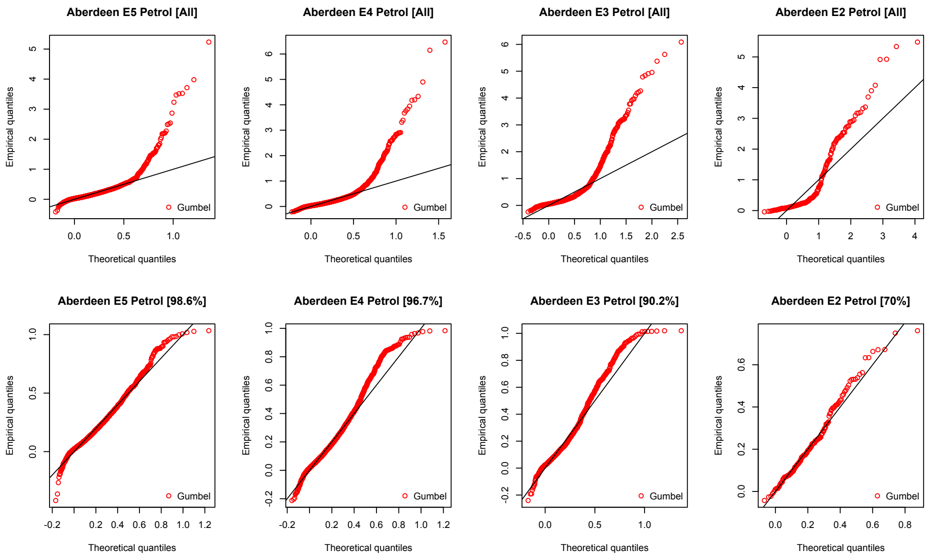

Figure 5.6: E5, E4, E3 and E2 Petrol NO_X emissions Q-Q Comparison plots for all and cut sections

sented alongside them. The criteria for the cut point was to create the closest possible straight line in a Q-Q plot. The results of the Q-Q plots are presented in Figure .

The model fits for the fleet subsections with the super highly emitting vehicles removed conforms in a much better way to the Gumbel distribution that they are fitted to. This analysis shows that with the appropriate removal of vehicles that are clearly not behaving in the same way as the vast majority of the fleet a model can still be fitted to the data. If the fleet is to be accurately modelled it is still important to understand how the final few percent of vehicles behave statistically. Figure 5.7 shows the density distribution of the total NO_X emission factor for these vehicles.

The shape of the tail of the distribution is similar to that of an exponential decay in the emission factor values in Euro 5, 4 and 3 however the distribution of Euro 2 super high emitters appears to be

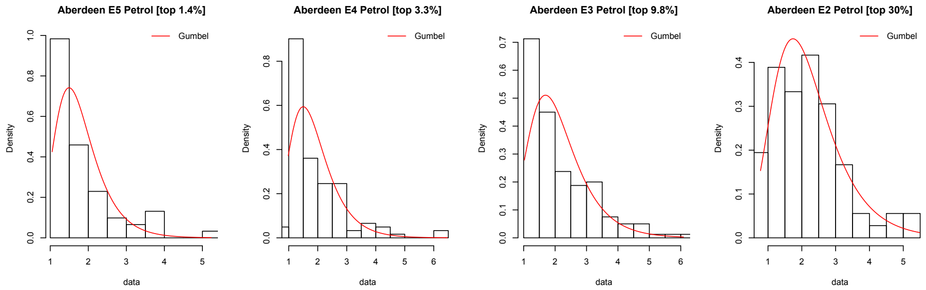

Figure 5.7: E5, E4, E3 and E2 Petrol Density Comparison plots for the off-model highest emitters

more uniform across the range. The cause of these super extreme values remains an open question. Given that the percentage of vehicles that exhibit these characteristics increases significantly with the age of the vehicle it is hypothesised that they are predominantly caused by ageing within the vehicle systems however the metadata associated with the vehicle measurements does not allow us to make this distinction. Borken-Kleefeld & Chen (2015) finds that vehicle mileage has little impact on the propensity for a vehicle to be in the highest quantile of emission so it may also be hypothesised that these observations are caused by sub-optimal catalyst operating conditions leading to either the vehicle switching off the catalyst, catalytic breakthrough (e.g Morgan *et al.*, 1973) ocuring or the catalytic converter operating under cold-start type conditions. Further investigation of these observations is undertaken in Section 5.4.2.

5.3 Applications to Urban Environment Driving Data

It has been demonstrated that using the Gumbel distribution to describe fleet vehicle emissions is a robust method for most of the fleet of passenger cars. This section will review a number of short applications of this method designed both to demonstrate the validity of this approach to describe real world vehicle emissions behaviour. This section will also begin to examine candidate hypotheses for the cause of the off-model behaviour observed for a small number of passenger cars.

5.3.1 The Quantitative Difference Between Petrol and Diesel Powered Vehicles

A simple test to demonstrate the use of the method to provide useful information about the NO emissions of a population is to quantify the difference between the petrol and diesel fleets. It is well known that older diesel powered vehicles emit more NO than their petrol powered counterparts however due the asymmetry in the data a simple mean and standard deviation model lacks the subtleties required to describe the distribution of measurements. To back this up a two-sample K-S test was applied to the data. The vehicles selected for this analysis were the sourced from the Aberdeen data-set as it is the most recently acquired set. Selection criteria was Euro 3 - 5 petrol and diesel powered passenger cars. Euro 6 vehicles are analysed in depth in Chapter 6. The vehicle subsets were fitted to a normal distribution function using the *fitdistrplus* package in R (Delignette-Muller & Dutang, 2015; R Core Team, 2015). The mean and standard deviation were extracted from the fit and a test sample of 1000 random numbers fitting this distribution were created. A p-value for K-S test was created and for both the Euro 5 and Euro 4 petrol vehicles $p < 2.2 \times 10^{-16}$ As such the hypothesis that these two data sets come from the same underlying distribution is discarded backing up the assertion made in Section 5.1. The same test is applied for a set of randomly distributed numbers following the Gumbel distribution of the on-model fleet. For Euro 5 vehicles $p = 0.06$ and for Euro 4 vehicles $p = 0.55$. Both of these values are high enough to accept the hypothesis that the observed data comes from the fitted distributions backing up the assertion made in Section 5.2. The considerably improved p statistics for the Gumbel distribution show that it accounts for the observed asymmetry and is suitable for describing these data in a more sophisticated way.

The Euro 2 sections of the fleet are not compared as a significant number of the petrol vehicles were observed to be off-model and there are a lot less of them. To investigate the difference between Euro 3, 4 and 5 class diesel vehicles operating in real driving conditions the data from the Aberdeen study (Section 3.4.4 was analysed. Total NO_X was calculated using Equation 3.5. As mentioned previously the off-model emitting top couple of percent of petrol vehicles cannot be modelled using the standard extreme value distribution so the on-model population is considered on it's own. The tools for analysing these vehicles described in Section 5.4.2 were not fully realised at the time of analysis. The trend towards older vehicles being more likely to be off-model suggests that there is some physical change going on within the vehicle that is causing this however using this data alone it cannot be shown what is causing the increase in poorly behaved vehicles with age. This is of concern because if the only factor is the vehicle age then the behaviour of the current generation of new petrol powered vehicles may begin to exhibit this behaviour characteristic in the future. In all cases the entire diesel fleet is used as it's members are well behaved across the entire range of emission factor values.

The vehicle emission factors are described in terms of the location parameter a and the scale parameter b. The location parameter corresponds to the modal value of the fleet and the scale parameter indicates the influence of the tail on the distribution. The parameters for each fleet subset are presented in Table 5.1. The errors presented alongside the parameter values represent the standard error as calculated by the distribution fitting algorithm. In the context of this research question a lower value for both the location parameter and the scale parameter is a better result as it relates to a low value with little variance.

Fleet Section	Location (a) [gkm^{-1}]	Scale (b)
Euro 5 Diesel	0.466 ± 0.005	0.360 ± 0.004
Euro 4 Diesel	0.542 ± 0.004	0.443 ± 0.008
Euro 3 Diesel	0.61 ± 0.02	0.443 ± 0.008
Euro 5 Petrol	0.113 ± 0.002	0.138 ± 0.002
Euro 4 Petrol	0.131 ± 0.003	0.162 ± 0.002
Euro 3 Petrol	0.210 ± 0.007	0.291 ± 0.006

Table 5.1: Quantitative comparison between petrol and diesel powered vehicles in Aberdeen

It is no surprise that petrol vehicles out perform diesel vehicles emitting less NO_X than their diesel counterparts however parameterising it in this way allows a direct comparison to be made using meaningful metrics. It can also be seen that across the fleet there has been very little real world improvement in NO_X emission factors between Euro 4 diesel vehicles and Euro 5 diesel vehicles. The modal peak for Euro 5 diesel passenger cars is 85% that of Euro 4 diesel passenger cars compared to the 72% decrease in limit values from Euro 4 to Euro 5. Prior to this analysis the lack of significant change between Euro 4 and 5 diesel vehicles was believed to be real based on average emission factors however it has been shown by this analysis that the NO_X emission factors of two subsections of the fleet behave in similar ways. This confirms observations made previously (Carslaw & Rhys-Tyler, 2013). Some improvement is observed from Euro 3 to Euro 5 however it is nowhere near as great as the difference in legislated type approval emission factor limit values has been. There is evidence of a decrease in emissions from older to newer Euro classes of petrol powered engines, suggesting that the introduction of lower limit values in subsequent Euro class legislation has been at least in part responsible for decreased emissions.

5.3.2 Impact of Vehicle Specific Power on Emission Factor Distribution

It has been shown in Carslaw *et al.* (2013) and Wyatt *et al.* (2014) that high VSP (Section 4.2.1) is related to high NO_X emissions. The remote sensing data presented in Section 5.2 shows that there are a significant number of vehicles who's emissions do not fit within the Gumbel distribution and are therefore off-model. The theoretical and measured quantiles of this data are not the same as would be expected if they were on-model. The emissions recorded for these vehicles are often significantly higher than would be expected if they were on-model. Given the assertion based on the literature that the high emission of NO_X is related to high VSP it is hypothesised that these off-model emitters is caused by excessively high VSP requirement of the vehicle as it was measured. To test this hypothesis the NO_X emission factor for each vehicle is subset into bins based on their observed VSP quantile for each measurement. If this hypothesis is correct then removal of the highest quantiles of VSP vehicles from the data set will result in the removal of the highest NO_X emitters too. Failure of this cutting of the data to remove the highest emitting vehicles will show that these off-model observations are not connected to the VSP and are caused by another factor, possibly a failed catalyst, a cold start, damage to the catalyst system or further external factors.

In this section this hypothesis will be tested using remote sensing data gathered in Cambridge and Sheffield as the range of different road gradients, a parameter in the VSP calculation, are available. The Sheffield and Cambridge petrol passenger car fleets are combined again and subsetted by their VSP as well as their Euro class with a Gumbel distribution fitted to each subset. The VSP subsets are based on the deciles whereby the total NO_X emission factors for each 10% of the fleet according to it's VSP can be compared with one another. The NO_X emission factors for the vehicles with the bottom 90% of the

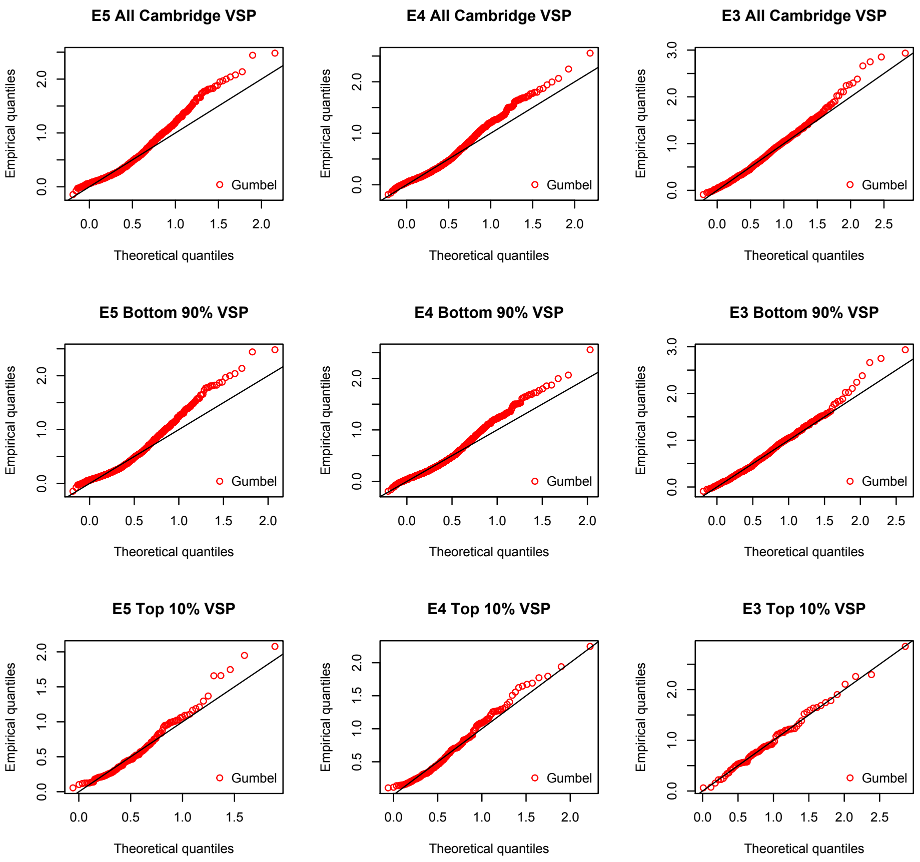

Figure 5.8: Analysis of petrol car emission factors of the bottom 90% and top 10% of VSP in Cambridge

observed VSP were compared to the whole fleet to determine whether or not the VSP influences the overall distribution. Plots for Cambridge and Sheffield are shown in Figures 5.8 and 5.9 respectively with the calculated values shown in Table 5.4

The results of this analysis show that vehicles that are being driven with heavy acceleration and high VSP, do not significantly impact the effectiveness of the Gumbel model to describe the distribution of emission factors across the fleet. There is small change, typically between $0.01 - 0.02 gkm^{-1}$, between the location parameter in the newer vehicles and extending to $0.02 - 0.04 gkm^{-1}$ for the older vehicles how-

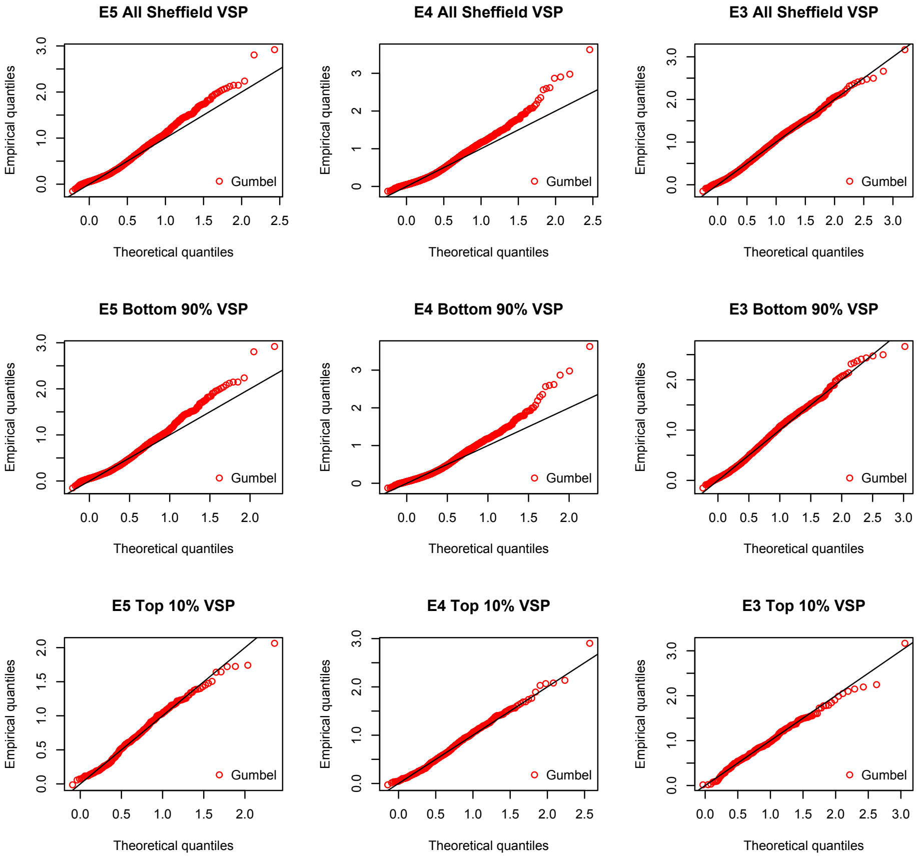

Figure 5.9: Analysis of petrol car emission factors from the bottom 90% and top 10% of VSP in Sheffield

Fleet	Location (a) [gkm^{-1}]	Scale (b)
E5 All Cambridge	0.296 ± 0.007	0.236 ± 0.005
E5 Cambridge 90%	0.286 ± 0.007	0.220 ± 0.006
E5 All Sheffield	0.309 ± 0.005	0.244 ± 0.004
E5 Sheffield 90%	0.294 ± 0.005	0.235 ± 0.004
E4 All Cambridge	0.270 ± 0.006	0.233 ± 0.005
E4 Cambridge 90%	0.251 ± 0.006	0.220 ± 0.004
E4 All Sheffield	0.281 ± 0.004	0.243 ± 0.003
E4 Sheffield 90%	0.259 ± 0.004	0.227 ± 0.003
E3 All Cambridge	0.44 ± 0.01	0.326 ± 0.009
E3 Cambridge 90%	0.41 ± 0.01	0.308 ± 0.010
E3 All Sheffield	0.452 ± 0.008	0.335 ± 0.006
E3 Sheffield 90%	0.428 ± 0.008	0.321 ± 0.006

Table 5.2: Quantitative comparison between the bottom 90% of the fleet and the whole fleet

ever given the standard error calculated by the fitting algorithm the fitted parameters are generally equivalent. The evidence is particularly clear in the Q-Q plots (Figure 5.9) where the same deviations by the highest emitters are observed in both the full and the cut data set. Furthermore vehicles with the top 10% VSP are still modelled by a Gumbel distribution albeit with different parameters (discussed in Section 5.4). The implication of this is that these vehicles are not being cut from the set because of their high VSP and therefore it can be asserted that the behaviour of the driver is not causing the departure from the distribution. This is a significant finding as it shows that it is not just the driving behaviour that leads to the observed distribution functions. Whilst the higher NO_X emission values causing the asymmetrical emission factor distribution are likely to be related to VSP they are likely to still be on-model with the off-model fraction not linked to VSP. The dependence of the emission factor on the VSP is folded into the Gumbel distribution. The cause of these off-model vehicles remains unknown however it remains more likely that they are caused by suboptimal operation of or damage to the emission reduction systems be this due to damage, age or the operation of the onboard control systems.

Given that prior work (Carslaw *et al.*, 2013; Wyatt *et al.*, 2014) shows that VSP impacts on the NO_X emission factor one final examination of the data is performed. The Euro 5 diesel powered passenger cars from Cambridge are split into their deciles and the deciles are analysed individually to determine whether or not improvements to the fleet model can be achieved this way. The results of this analysis are presented in Figure 5.10.

This analysis shows that no additional convergence to the model is observed by separately analysing the VSP deciles and furthermore this fine tuning of the model may result in the addition of more off-model extreme values at lower deciles.

There are two types of potential causes of the off-model results observed throughout this section. Either they are because of cold starts or they are because of failed catalysts for whatever specific reason the failure occurred. It is unreasonable to assume that there is a higher cold start percentage across different euro classes so it is hypothesised that the percentile that are removed in the Euro 5 for Aberdeen (1.4% Figure 5.2.1) relate to the cold starts as these catalysts have had the least time to fail due to ageing. If the assumption that the rest of the vehicle fleet subsections have the same percentage of cold starts as the Euro 5 percentage then the difference would be the number of vehicles with a failed catalyst. The projected failed percentage rate for catalysts is therefore 1.9% for Euro 4, 8.4% for Euro 3 and 28.6% for Euro 2. The fleet size for Euro 1 is too small to get an accurate result. These percentiles can be modelled as part of an attempt to predict the impact that these vehicles will have on the total emission

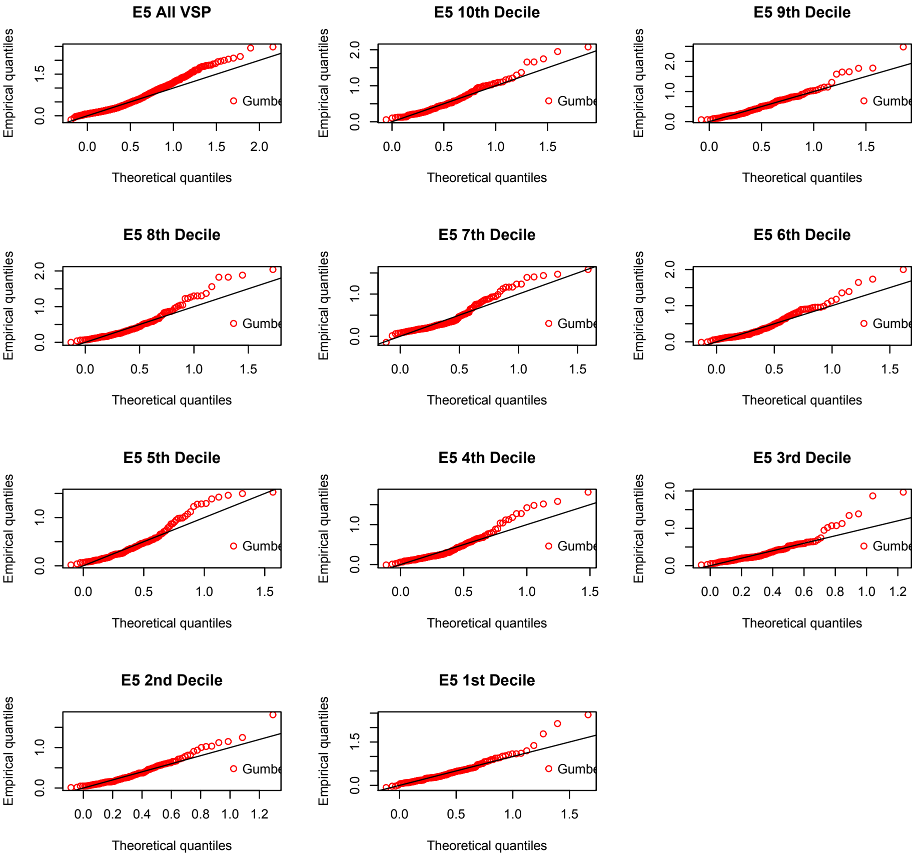

Figure 5.10: Analysis by VSP decile of the total NO_X emission factors of Euro 5 diesels in Cambridge

inventory for that vehicle subset. Cold start candidates are analysed in Section 5.3.4. The implications of this result are useful because it means that assertions may be made about the catalyst failure rate in older vehicles. There are wide ranging implications on real world policies relating to catalyst degradation fraction so being able to attempt to quantify the number of vehicles or percentage of vehicles of a fleet subset that are fitted with a poorly operating catalyst is important.

5.3.3 Impact of Site Selection on Distribution Function Parameters

The selection of the site is important in the creation of an RSD study as good site selection leads to a better data capture success rate. Over the course of the Sheffield and Cambridge studies (Sections 3.4.1 and 3.4.2 respectively) ten different sites were measured for two days each. The sites featured a variation in road gradients and driving conditions from relatively steep urban areas to much flatter suburban areas. Understanding the differences in emissions from site to site allows for a more detailed description of the sources of vehicle emissions over a network as a whole. It may help to unpick some of the unanswered questions that the previous emission factor models have uncovered in the behaviour of the fleet as a whole. Given that VSP has been previously related to NO_X emissions (Carslaw *et al.*, 2013) sites that have a steeper gradient might be expected to have a higher peak NO_X emission and a greater number of on-model high emitters.

To test this hypothesis the Sheffield and Cambridge datasets were subset into their individual locations and the two days measured at each site were combined into one subset. Variance between the initial and final environmental conditions (Section 4.2.2) were more varied than the intra-day variance so combining the two days measurements into one data set can be done. The vehicles were further subset into their fuel and Euro class. Diesel vehicles were the only fuel type considered as their emissions have been shown to be considerably higher than those of petrol engines (Section 5.3.1. Total NO_X emission factors are calculated for each vehicle using Equation 3.5 and uplift factors for vehicle CO_2 emissions are sourced from Tietge *et al.* (2015). The Q-Q plots from the fitted distributions are shown in Figures 5.12 and 5.14 for Euro 5 and Euro 4 diesels respectively. The maximum likelihood best fit parameters are shown in Tables: 5.3 and 5.4 for Euro 5 and Euro 4 diesels respectively.

Euro Site	Location (a) [gkm^{-1}]	Scale (b)
E5 Cambridge (All, $n = 2404$)	0.296 ± 0.007	0.236 ± 0.005
E5 East Road ($n = 842$)	0.28 ± 0.01	0.224 ± 0.009
E5 Elizabeth Way ($n = 331$)	0.29 ± 0.02	0.22 ± 0.01
E5 Elizabeth Way (Humberstone, $n = 635$)	0.30 ± 0.01	0.25 ± 0.01
E5 King Street ($n = 63$)	0.31 ± 0.05	0.28 ± 0.04
E5 Newmarket Road ($n = 533$)	0.32 ± 0.01	0.24 ± 0.01
E5 Sheffield (All, $n = 4890$)	0.309 ± 0.005	0.244 ± 0.004
E5 Asline Road ($n = 446$)	0.36 ± 0.02	0.30 ± 0.02
E5 Attercliff Centre ($n = 1538$)	0.301 ± 0.008	0.223 ± 0.006
E5 Eyre Street ($n = 589$)	0.33 ± 0.02	0.28 ± 0.01
E5 Prince of Wales Road ($n = 937$)	0.31 ± 0.01	0.230 ± 0.008
E5 Western Bank ($n = 1380$)	0.249 ± 0.009	0.248 ± 0.007

Table 5.3: Quantitative comparison between different sites across Sheffield and Cambridge for Euro 5 Diesel passenger cars

Euro Site	Location (a) [gkm^{-1}]	Scale (b)
E4 Cambridge (All, $n = 3986$)	0.270 ± 0.006	0.223 ± 0.005
E4 East Road ($n = 1257$)	0.260 ± 0.009	0.219 ± 0.007
E4 Elizabeth Way ($n = 572$)	0.24 ± 0.01	0.21 ± 0.01
E4 Elizabeth Way (Humberstone, $n = 1144$)	0.27 ± 0.01	0.233 ± 0.009
E4 King Street ($n = 158$)	0.30 ± 0.03	0.27 ± 0.03
E4 Newmarket Road ($n = 855$)	0.30 ± 0.01	0.26 ± 0.01
E4 Sheffield (All, $n = 6822$)	0.281 ± 0.004	0.243 ± 0.003
E4 Asline Road ($n = 706$)	0.30 ± 0.01	0.27 ± 0.01
E4 Attercliff Centre ($n = 1956$)	0.284 ± 0.007	0.239 ± 0.006
E4 Eyre Street ($n = 1053$)	0.35 ± 0.01	0.29 ± 0.01
E4 Prince of Wales Road ($n = 1353$)	0.295 ± 0.009	0.225 ± 0.007
E4 Western Bank ($n = 1754$)	0.229 ± 0.002	0.216 ± 0.006

Table 5.4: Quantitative comparison between different sites across Sheffield and Cambridge for Euro 4 Diesel passenger cars

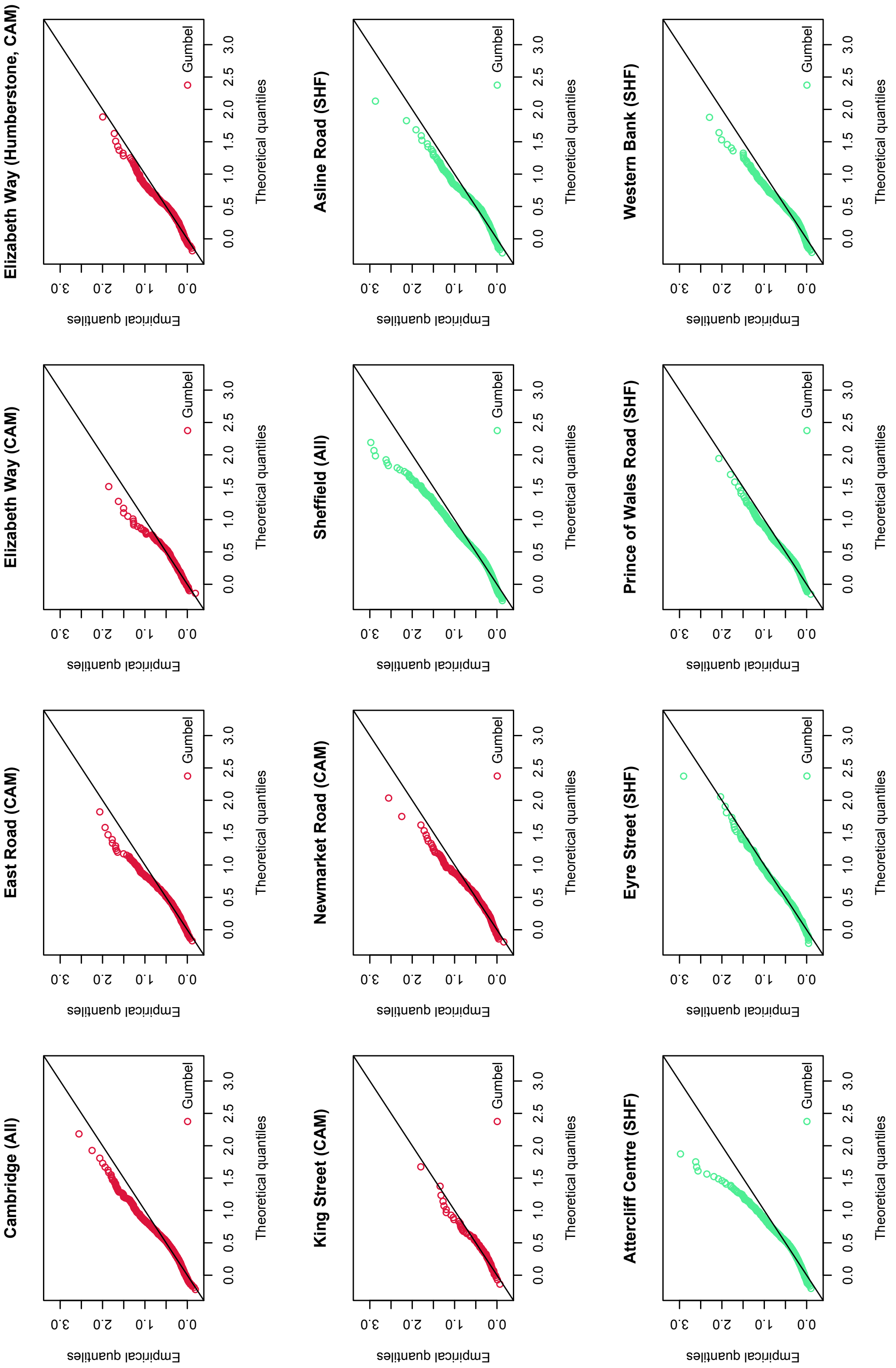

Figure 5.11: Cross site Q-Q plots of E5 diesel cars

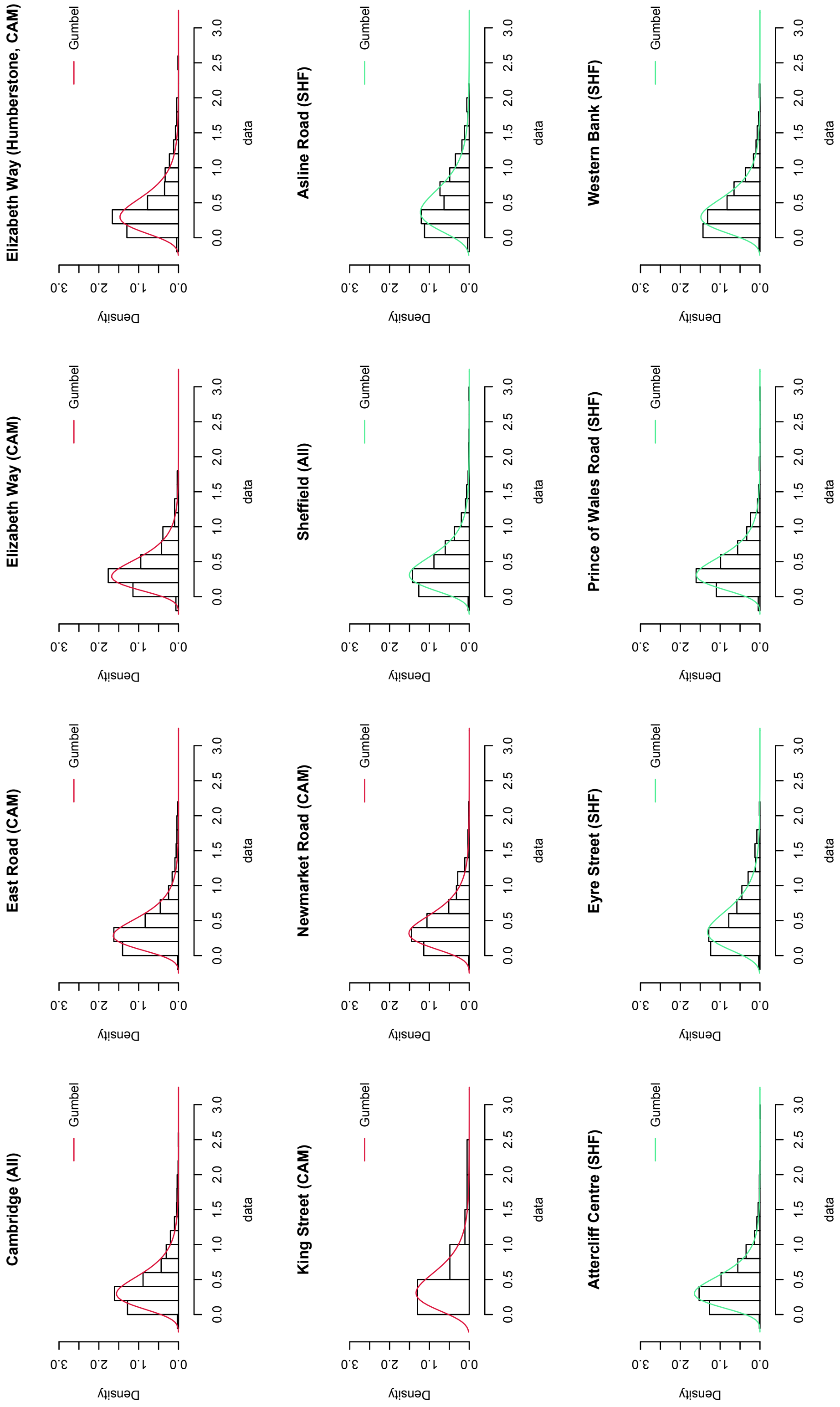

Figure 5.12: Cross site Density plots of E5 diesel cars

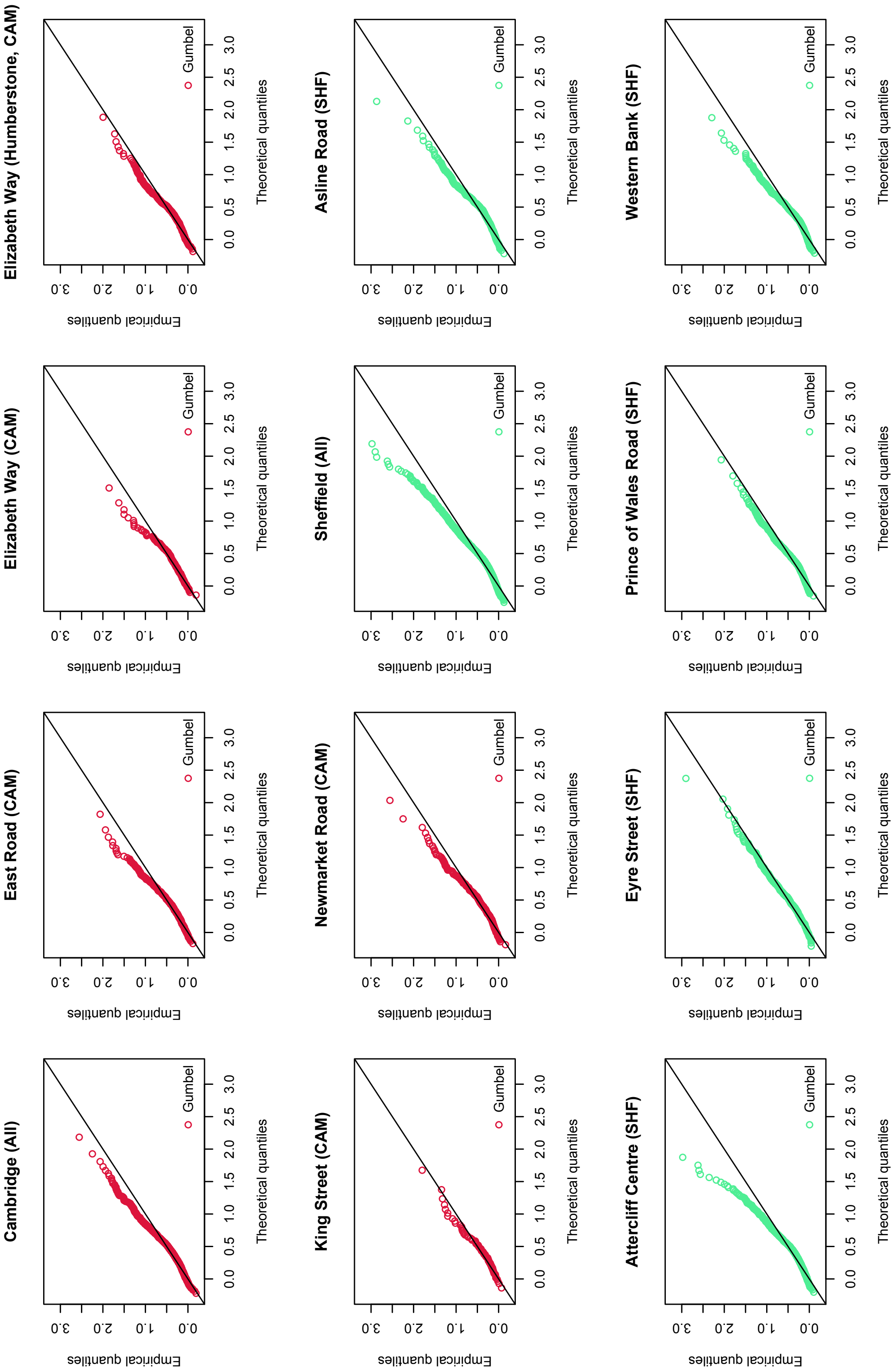

Figure 5.13: Cross site Q-Q plots of E4 diesel cars

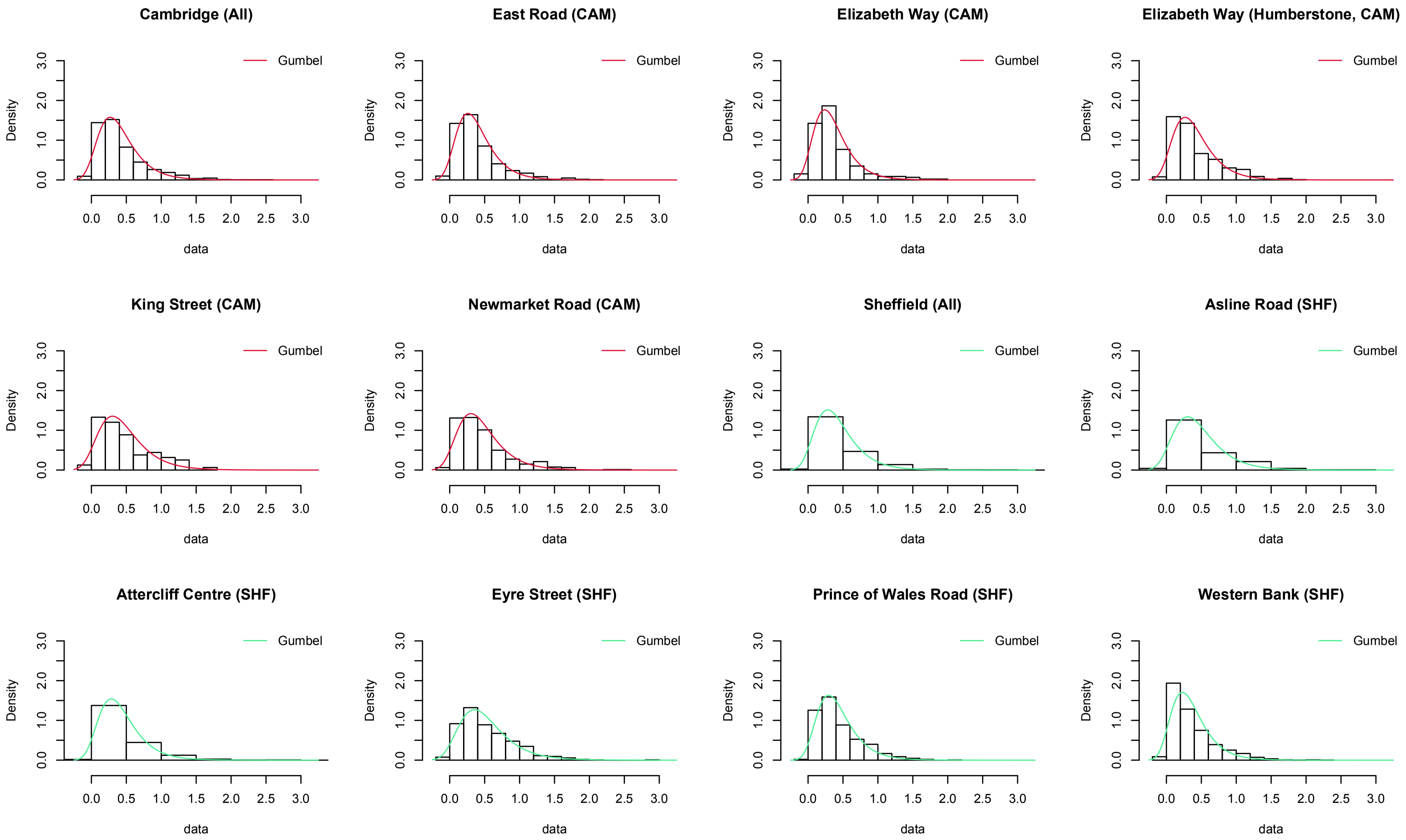

Figure 5.14: Cross site Density plots of E4 diesel cars

The results of the cross city analysis show that there is a measurable difference between Cambridge and Sheffield. Cambridge shows a lower location parameter and a tighter spread of emissions than Sheffield in both the Euro 4 diesel and the Euro 5 diesel vehicle population. The differences are not large and the reasons for this are not immediately clear from the data in it's current form. It is likely that the lack of significantly hilly areas requiring less engine load at the time of measurement in Cambridge are responsible at least in part however factors relating to the vehicle such as mechanical maintenance, catalyst ageing and other regional factors may further influence this outcome.

The results of the cross site analysis show that for Cambridge, which is almost completely flat, the parameters, given the standard error, are relatively consistent across the selection of sites. The one site with significant topographical difference to this is Newmarket Road. At the Newmarket Road site the RSD was set up approaching a bridge over the railway line. The additional engine load required at this very short section of higher than normal road gradient for Cambridge may be the cause for the slight increase in NO_X emission of the Euro 4 vehicles at this point. The parameters estimated at King Street are subject to higher errors. The King Street site (Section 3.4.2) was a low traffic flow site and not ideal for RSD survey but included to be representative of the historical centre of Cambridge.

The parameters for the Sheffield sites vary a small amount depending on their location. The potential for different sites in Sheffield is greater as it is not as flat as Cambridge however the site that had consistently the lowest instantaneous total NO_X emission factors was the Western Bank site. This was located at the top of a relatively steep climb. Eyre Street, Prince of Wales Road and Asline Road on the other hand were located in the City Centre and a quiet suburb respectively. A potential reason, although not testable using remote sensing data alone, for this result is that the vehicles approaching the

RSD have just completed a significant period of higher than normal engine loading and therefore all the catalyst systems are running at a more optimal temperature than they would be if they had been sat in slow moving traffic in the city centre or only just started their journey from an urban location. The higher peak emissions in Eyre street may also be due to the presence of a larger number of taxis and private hire vehicles. The difference between taxis and privately owned vehicles is explored in Section 7.4.

The cross site analysis also shows that some sites are more susceptible to off-model NO_X emitters. This is especially noticeable in the Sheffield data set where the Attercliff Centre and Prince of Wales Road population shows a higher number of off-model emitters for Euro 5 diesel cars compared to the rest of the sites and Attercliff Centre also has a higher number of off-model Euro 4 vehicles than the other sites. This has implications for modelling the emission factors as these off-model vehicles may only be driven in a small spatial area. Modelling of this area by it's self may lead to an underestimation of their contribution or may lead to an overestimation if their probability of occurring decreases when taking the whole network into account. If they are introduced into areas that they would not normally be found in they could distort the result of the model.

The conclusions to be drawn from this investigation is that whilst the site selection can influence the total NO_X emission factors measured by the RSD, especially for older vehicles, the variation between sites may be also be influenced by the context of the site in the network as a whole. This result is not particularly surprising given the complicated nature of the emission control systems found in modern diesel vehicles. The difference between Euro 4 and Euro 5 class vehicles as they relate to different sites has shown both to be sensitive to site selection. The only major departure from this result is the Eyre

Street Euro 4 diesel cars which appear to be significantly higher than the other measurements.

5.3.4 Impact of Cold Starts on Emission Distribution

It is hypothesised that vehicles that exhibit off-model $NO : CO_2$ values as well as a large CO emission are candidates for vehicles with cold starts. A cold catalyst would cause an increase in NO emission as well as an increase in emission of CO and HC from the engine (Bielaczyc *et al.*, 2014; Dardiotis *et al.*, 2013; Singh & Prasad, 2014; Weilenmann *et al.*, 2005). The remote sensing device measured CO alongside NO (Section 2.6.2) so a comparison of these maximum values is possible. It is difficult to estimate the number of cold start vehicles using remote sensing devices without prior knowledge of the vehicles' behaviour which is not available on on-road studies. An attempt to estimate the number of cold starts present on the roads would be a useful tool for people attempting to model vehicle emissions network-wide. The $NO : CO_2$ and $CO : CO_2$ ratios from the Euro 5 petrol Aberdeen dataset were compared. These vehicles have number of off-model $NO : CO_2$ measurements and might be considered too new for a large number of failed catalysts. It is therefore hypothesised that these vehicles that are off-model are likely to be cold-start candidates. It was observed that in both cases the 98^{th} percentile for both the $CO : CO_2$ and $NO : CO_2$ ratios are a significant departure from the rest of the fleet. These measurements were isolated in both cases and vehicles that were present in both sets were noted. Of 88 vehicles in the top 98% of the $NO : CO_2$ measurements 16 of these were also present in the top 98% of the $CO : CO_2$ measurements, a fraction of 18% of the top 2% or 0.36% or approximately one-in-three-hundred of the total fleet. At the time of writing this result cannot be validated however tests performed that measure the temperature of the plume or isolate vehicles that have undergone a long heat soak could confirm or refute this assertion. The rest of the vehicles in the top 2% of vehicles are emitting highly for other reasons and will be discussed in Section 5.4.2.

This fraction is very low however the methodology remains plausible. Dedicated tests designed to investigate cold starts are required to better understand the effect. It is proposed that the remote sensing device is set up on the exit lane of a long stay car park such as those found at airports. The vehicles measured at this point would all have undergone a long period of inactivity and had a similar heat soak. Measuring these vehicles and comparing them to the regular fleet would be the best way to understand the impact of cold starts. If a significant excess in emissions was observed these results could be used to identify cold start vehicles on the road. If no real difference is observed it would suggest that the contribution of cold start emissions in urban environments is very low. The implication of this result are that the contribution to the off-model fraction of NO_X emission by vehicles operating under cold start conditions are low in this dataset. It also means that these off-model vehicles are generating their excess pollution through a different mode. From a modelling point of view this means that the process for describing off-model vehicles is less complex as only the failed catalyst distributions need to be considered.

5.3.5 Urban Environment Summary

It has been shown that the emission factors of on-model vehicle subsets largely follow the Gumbel distribution and that it can be used to mathematically model the vehicle fleet. It has also been shown that the normal distribution function is not suitable for describing the behaviour of the fleet as a whole. Various other distribution functions have been tested and shown to be inferior to the Gumbel distribution from both statistical and practical viewpoints. The hypothesis was initially tested on a data set measured soon after an extended period of engine load (Zurich dataset) to attempt to eliminate any rogue measurements that might be caused by vehicles whose emission control systems were running suboptimally in an urban environment. Once it was proved that the Gumbel distribution was a plausible distribution to analyse the fleet the analysis was continued using datasets taken in urban environments in Cambridge, Sheffield and Aberdeen.

In the hot running data some rogue vehicles were observed that were emitting higher than would be expected even using the Gumbel extreme value distribution. The cause of these super high measurements remain unknown however analysis of new vehicles compared to old suggests that a higher number are present in the older fleet so it is likely that the cause is related to ageing, poisoning or otherwise damaging of the emissions control system. This hypothesis remains to be tested and requires additional data to be collected. In urban driving environments more off-model super high emitting measurements are observed than in the hot running environment. This suggests that vehicles are running sub-optimally when driven in an urban environment. It may be that in some cases higher engine load and additional time is required to sufficiently activate the catalyst systems or the vehicle emissions controls are being switched off due to a range of local environment parameters not meeting the criteria for activation or due to defeat device systems. The EC Directives (EC, 2007b) allow for vehicle emissions controls to be relaxed or switched off to protect the engine at cold temperatures as shown by Emissions Analytics (Telegraph, 2016) that manufacturers are using this option. The remote sensing device cannot assess the status of the onboard emissions control systems however further work can be done to assess the impact of temperature on these results.

A number of potentially key factors that could influence the total NO_X emission factors of vehicles as measured by a Remote Sensing

Device have been examined using the new mathematical framework. These results have implications in the derivation of a model to analyse the impact of potential interventions on vehicle emissions using this framework. Firstly the difference between petrol and diesel vehicles has been tested and it has been confirmed that petrol powered vehicles emit considerably less total NO_X than their diesel counterparts. This result is not new however the model of the fleet that has been created now allows for quantitative statements to be made that fit the observed data better than a simple mean and standard deviation analysis.

The impact of vehicle specific power on the distribution of total NO_X emission factors has been analysed and shown to not be a significant factor when fit parameters are being calculated. Whilst it is known that VSP influences NO_X emissions this result suggests that any VSP dependence is statistically accounted for when the distribution function is fitted to the fleet. The fleet is clearly two typed with the majority of the vehicles fitting in the regime defined by the Gumbel distribution with a small subsection, numbering only a few to a few tens of percent depending on the age of the vehicle age and fuel type being off-model. It has been hypothesised that these measurements are caused by catalyst failure in older vehicles and cold starts in newer vehicles.

The impact of the location on vehicles has also been tested across 10 locations in 2 different cities in the UK. It was hypothesised that sites with high gradient would produce higher emissions factors and that this might introduce a bias when the emission factors are extrapolated across the whole network. Analysis of the remote sensing device data has shown that variation between the location of test sites does exist however similar sites in different contexts will show statistically significant differences. The context of the site, the approach and possibly the different types of vehicles present in it, is of greater importance to the emission factor measurement than the site it's self.

5.4 Creating A Model Using Extreme Values

The first step towards modelling something is to be able to describe it mathematically. It has been shown that both the Gumbel distribution and the gamma distribution realistically represents a large section of the vehicle fleet with the Gumbel distribution proving to be a more useable framework with fit parameters that are better related to real features. Various tests and limitations have been performed on the Gumbel distribution to assess its validity in describing the emission factors of passenger cars driving in an urban environment. The Gumbel distribution has been shown to be successful at describing all but the most highly emitting vehicles. It is likely that these vehicles possess some characteristic that is atypical of the fleet as a whole but nonetheless needs to be recognised. Modelling the fleet using the Gumbel distribution along with the caveat of the super high emitters is therefore a valid mathematical model for the passenger car fleet as a whole.

A robust mathematical model will help assess the effectiveness of policy interventions designed to reduce total NO_X concentrations in urban environments by providing a more realistic quantitative value for potential reductions. The model should be able to give accurate predictions based on euro class and fuel type. Given better initial information it is hoped that appropriate use of this model will allow policy makers to make better decisions about how to implement low emission zones (LEZs) and ultra-low emission zones (ULEZ's) in areas where NO_X concentrations are especially high.

5.4.1 Model Description

The model is an empirically derived description of an in-situ fleet. The first step to creating such a model is to measure the fleet as it currently is directly using a remote sensing survey and create a database of current emission factors. Whilst older databases can be substituted at this stage the bespoke nature of the model will be impacted and the efficacy of the results will be reduced. It is recommended that vehicle emission factors are extrapolated across the network using the vehicle tracking and PHEM method described in Section 2.5.3 as this step will mean the results obtained are more representative of the real driving environment encountered by vehicles driving on the Aberdeen road network.

When a complete dataset has been collected the next step is to subset the vehicles into appropriate groups. The work done in Section 5.3 shows that appropriate subsections are fuel type and euro class. At this stage it is important to consider the impact of super high emitting vehicles and how they should be accounted for. These subsections have a Gumbel distribution function fitted to them and the model parameters are stored in a matrix referred to as the Location-Scale matrix (LS matrix). Along with distribution function parameters the fleet must also be described numerically. This is achieved by assessing the percentage of the total fleet that each subset represents. Once the vehicle type distribution and the emission factor distribution have been calculated the fleet can be statistically re-sampled.

The fleet is resampled using a random number 'Monte Carlo' method to determine the Euro class and fuel type of the vehicle and a fleet of new vehicles is created based on user chosen parameters. This new vehicle is assigned an emission factor randomly from a Gumbel distribution that with matching location and scale parameters from the LS matrix derived from the real data for it's subset. The process is repeated until a full fleet of vehicles has been created to the specification of the user. The full fleet and it's total NO_X emission factors have now been created and can be analysed in which ever way the user requires. The process of fleet generation should be repeated as many times as possible with the aggregation of all computationally derived fleets used rather than a single case.

5.4.2 Off-Model Measurements

Exponential Tail Hypothesis

For the vast majority of vehicles the Gumbel distribution realistically describes the behaviour of a fleet subset as a whole however in most subsets of the vehicle fleet there are vehicles who's emission does not fit the mathematical model as it currently stands. To accurately describe each subset and it's contribution to the total emissions these off-model vehicles must be accounted for.

At the point of calibration of the model the user must identify the percentile where the vehicles stop behaving in line with the model and start exhibiting characteristics of super highly emitting vehicles. Accessing this fraction is achieved by fine tuning the quantile cut from the Q-Q plot and is described in Section 5.2.1. Figure 6.5 shows the Euro 5 diesel fleet for Aberdeen with various top percentiles removed. For example the 99% plot is the fleet with the top 1% removed. The cut percentile is optimal when the predicted and theoretical quantiles best align represented by the straight line in a Q-Q plot. The optimal cut is made by adjusting the cut percentile to get a resulting distribution that best fits this line. In this example a case can be made for setting the percentile where the theoretical quantiles and the empirical quantiles best match up is at either 95% or 96%. The percentage point represented by ξ_{fleet} is expressed in the decimal form so $\xi_{95\%} = 0.95$. Two treatments of the off-model vehicles are proposed and tested in this section. The exponential tail model where the tail is modelled as an exponential function with parameters fine tuned to fit the data and the failed catalyst model where the vehicles in the off model fraction are modelled using an older class of vehicle, Euro 0 for petrol and Euro 5 for diesel Euro 6.

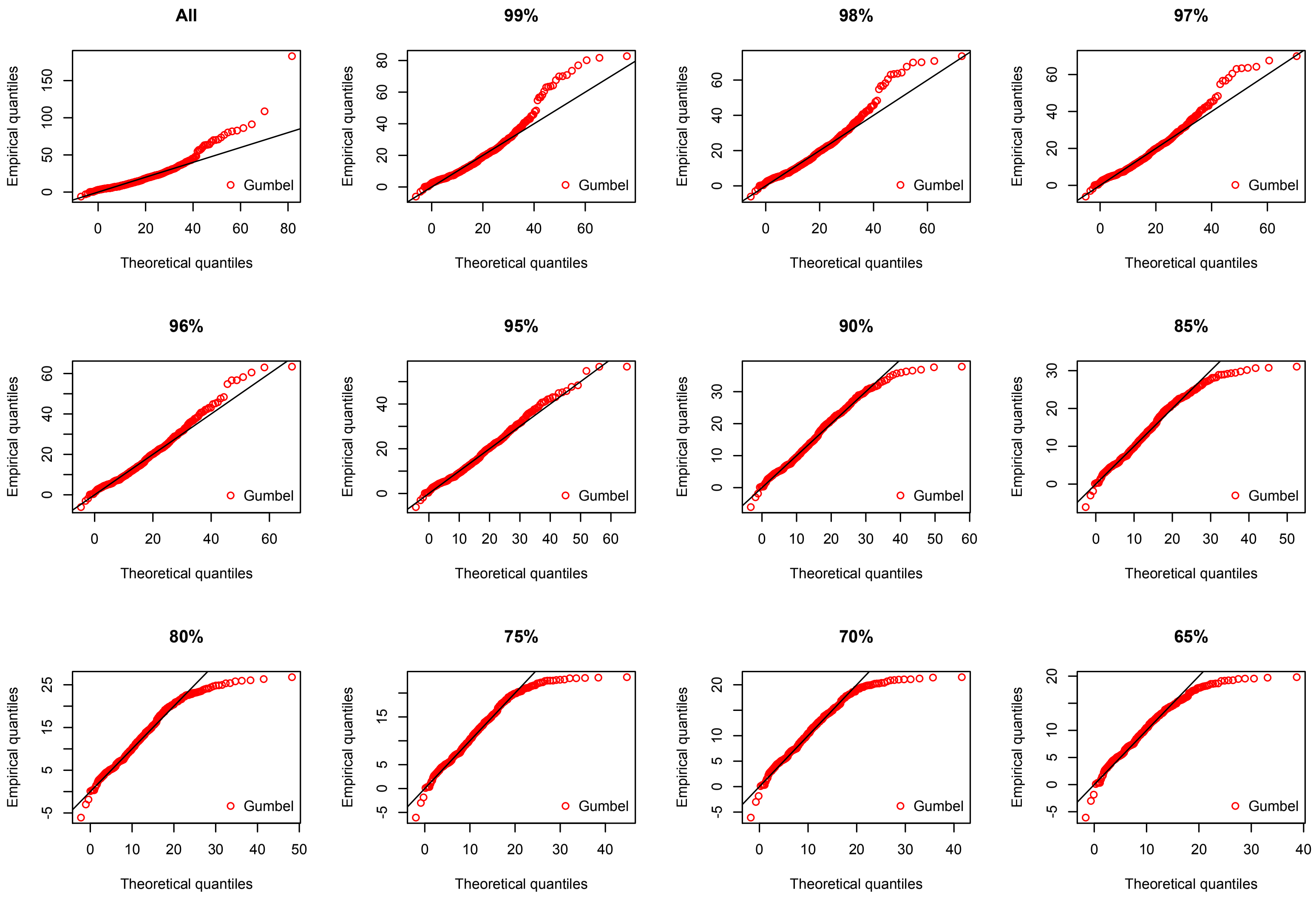

Figure 5.15: Q-Q plot by percentile of the E5 Diesel Aberdeen fleet

After the limiting point has been determined a second Monte-Carlo random number is generated between $0 - 1$. If the number is less than the ξ_{fleet} then it's emission ratio is resampled from the Gumbel distribution representative of it's subset. If it is greater than ξ_{fleet} and the exponential tail solution is to be used the emission ratio is generated by exploiting the exponential nature of the top n percentiles of the distribution as observed in Figure

$$\phi_{NO-shev} = \phi_{NO-q-turn} \times \eta_{exp} \tag{5.5}$$

This approximation is a non-elegant solution to the problem of accounting for these vehicles however it will be shown that given careful calibration of the rate parameters in this equation plausible approximations of the fleet emissions factors as a whole can be achieved. This model makes no attempt to understand the causes of these super highly emitting vehicles, instead it accepts that they are there and attempts to pragmatically assess their impact on the total NO_X emission by the fleet as a whole. Data beyond that collected by remote sensing devices is required to fully understand the cause and impact of these vehicles and that is beyond the scope of this thesis but remains an open question for future research.

Failed Catalyst Hypothesis

The Exponential Tail Hypothesis is unsettling because it requires a lot of fine tuning. Fine tuning is not desirable in scientific endeavour (Lightman, 1993) so a hypothesis that can be linked to a physical process is preferable. A further cause of the off-model vehicle behaviour could be due to failed or otherwise non-functioning catalyst systems. Whilst difficult to prove directly without additional data Section 5.4.2 shows attempts to circumstantially prove this by using both the exponential method and also by modelling the off-model fraction as Euro 0 vehicles for petrol vehicles and as Euro 5 vehicles for diesel vehicles. This is in line with the practice currently undertaken by NAEI (2012).

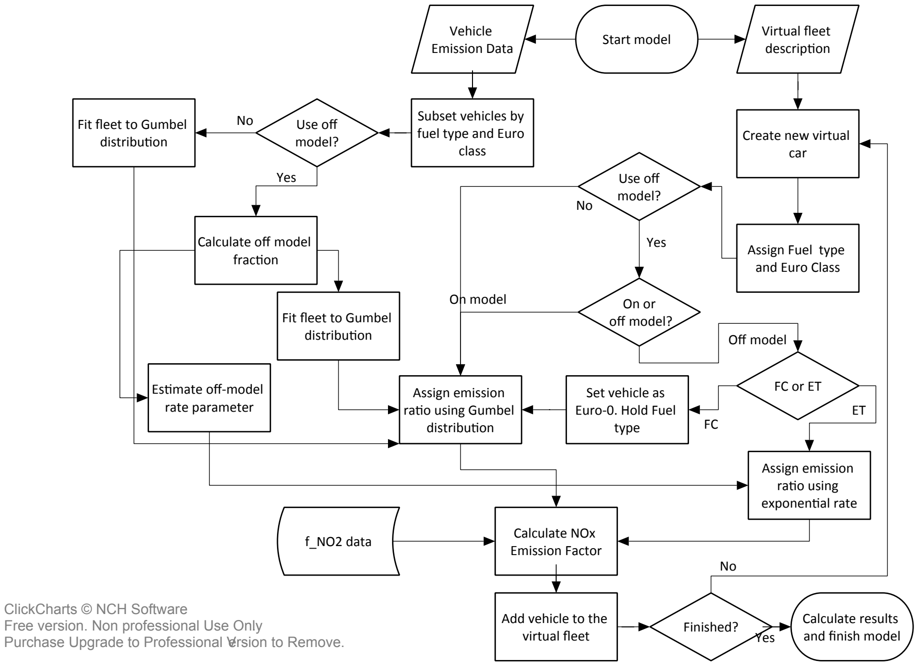

Figure 5.16: Flow chart for model

Hypothesis Testing

A flow chart for the proposed hypothesis testing model is shown in Figure 5.16. In this section both the off model hypotheses are tested.

To test the effectiveness of these hypotheses eleven simulations were conducted using the Aberdeen fleet distributions measured using the Remote Sensing Device and plotted next to the real data for comparison. The Euro 4, 5 and 6 petrol and Euro 6 diesel fleets were tested as they are the most relevant to ultra-low emission zones. The distribution of $NO : CO_2$ emission factors for the 11 simulated fleets and the observed fleet are shown in Figures 5.17, 5.18, 5.19 and 5.20. The real data is plotted in purple and the simulated data are plotted in red. The fitted model for each data set is overlaid in red.

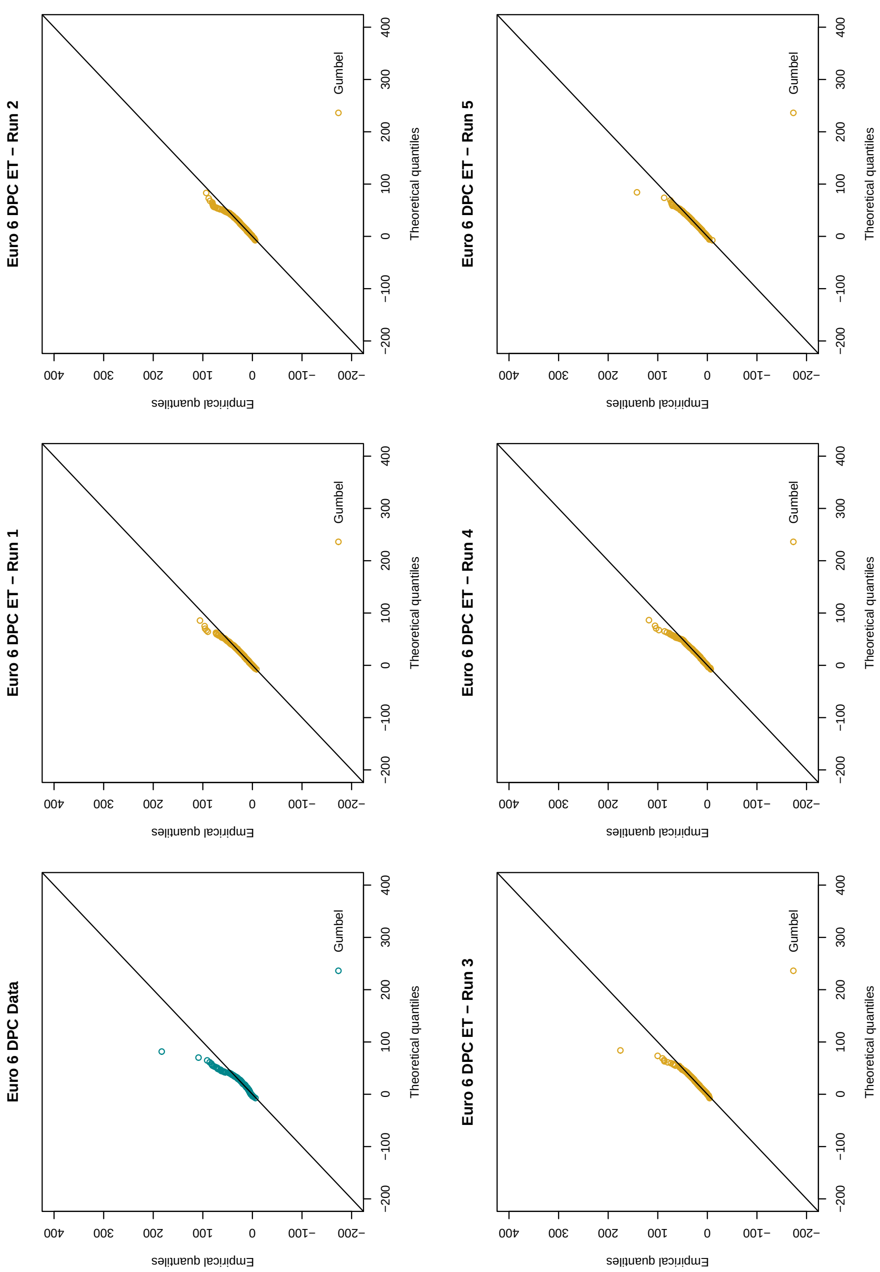

Figure 5.17: QQ Plot of Euro 6 diesel passenger cars modelled using exponential tail

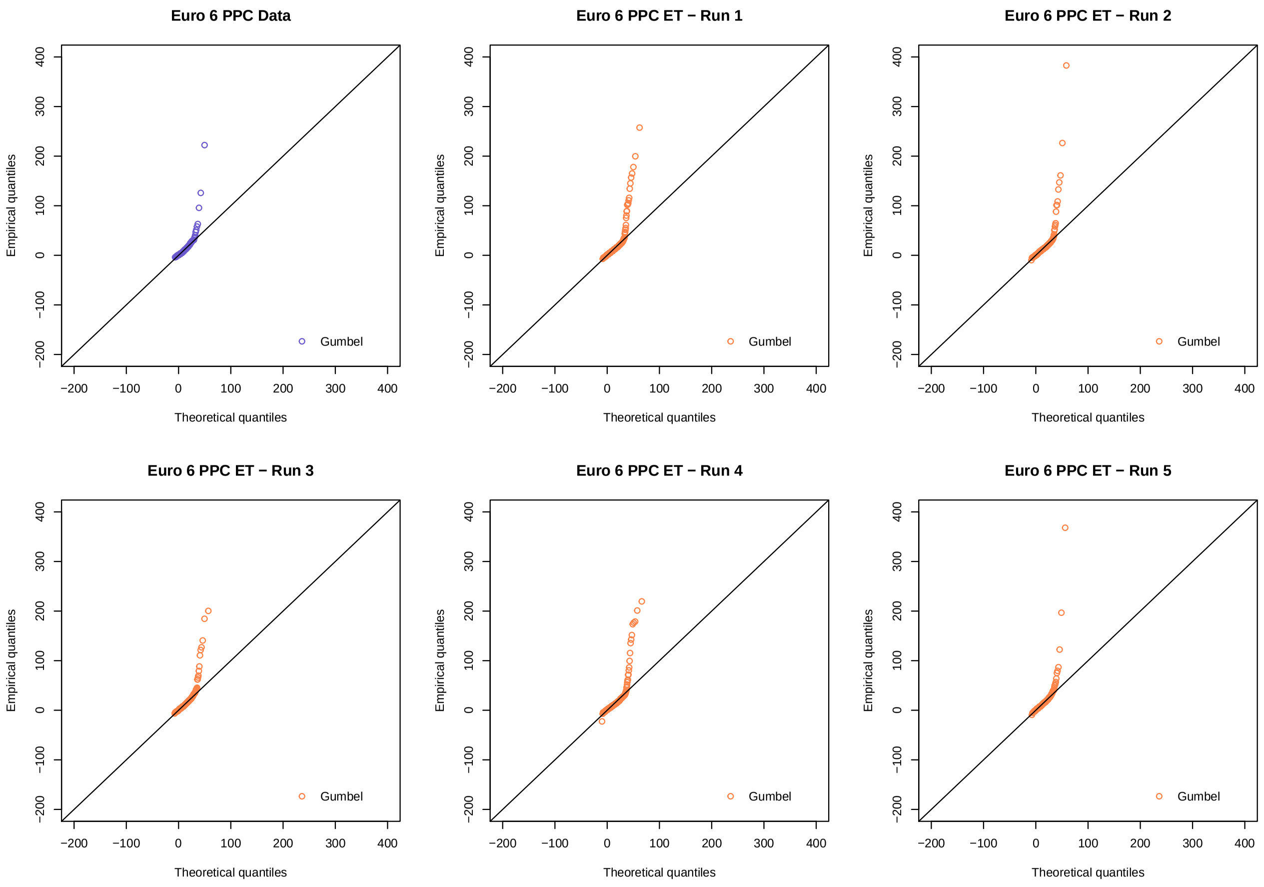

Figure 5.18: QQ Plot of Euro 6 petrol passenger cars modelled using exponential taill

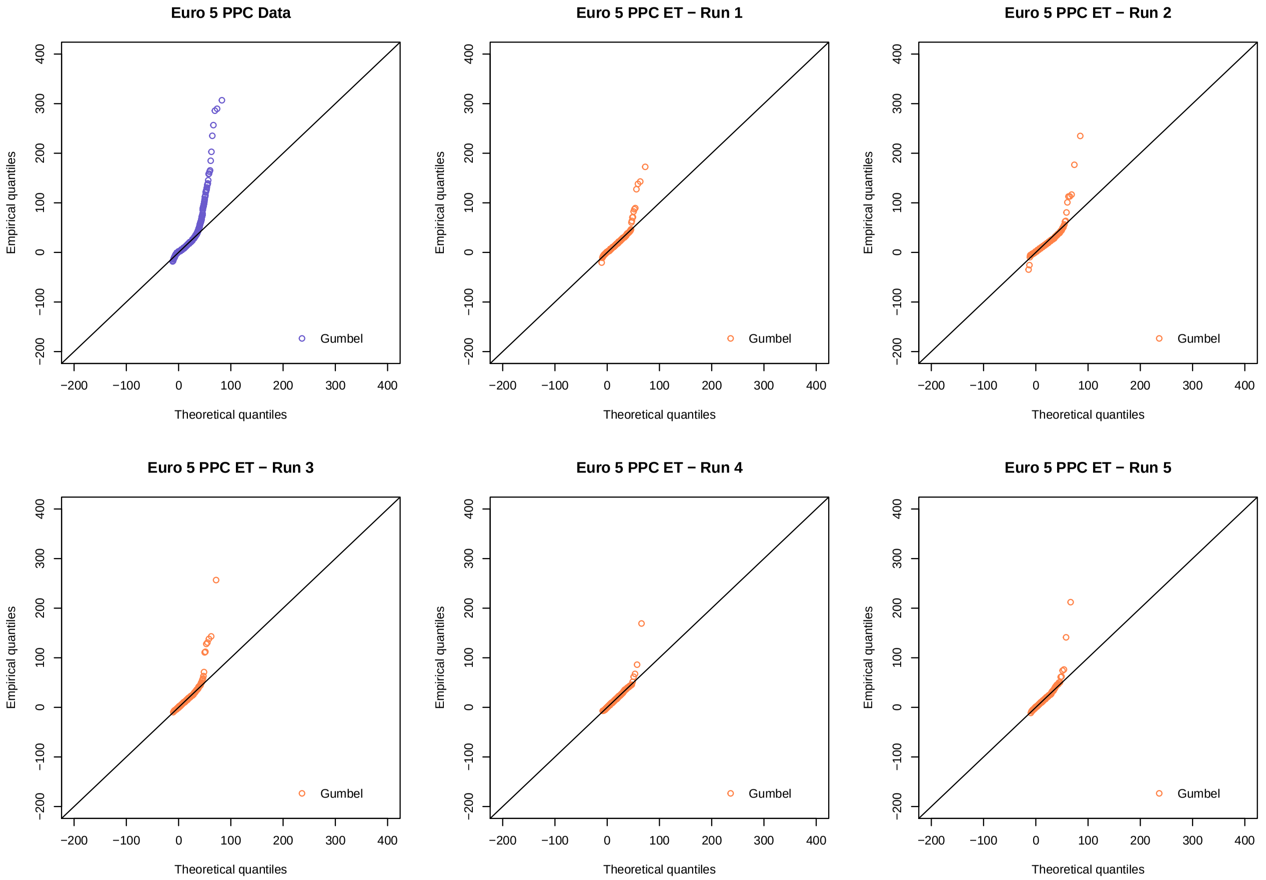

Figure 5.19: QQ Plot of Euro 5 petrol passenger cars modelled using exponential tail

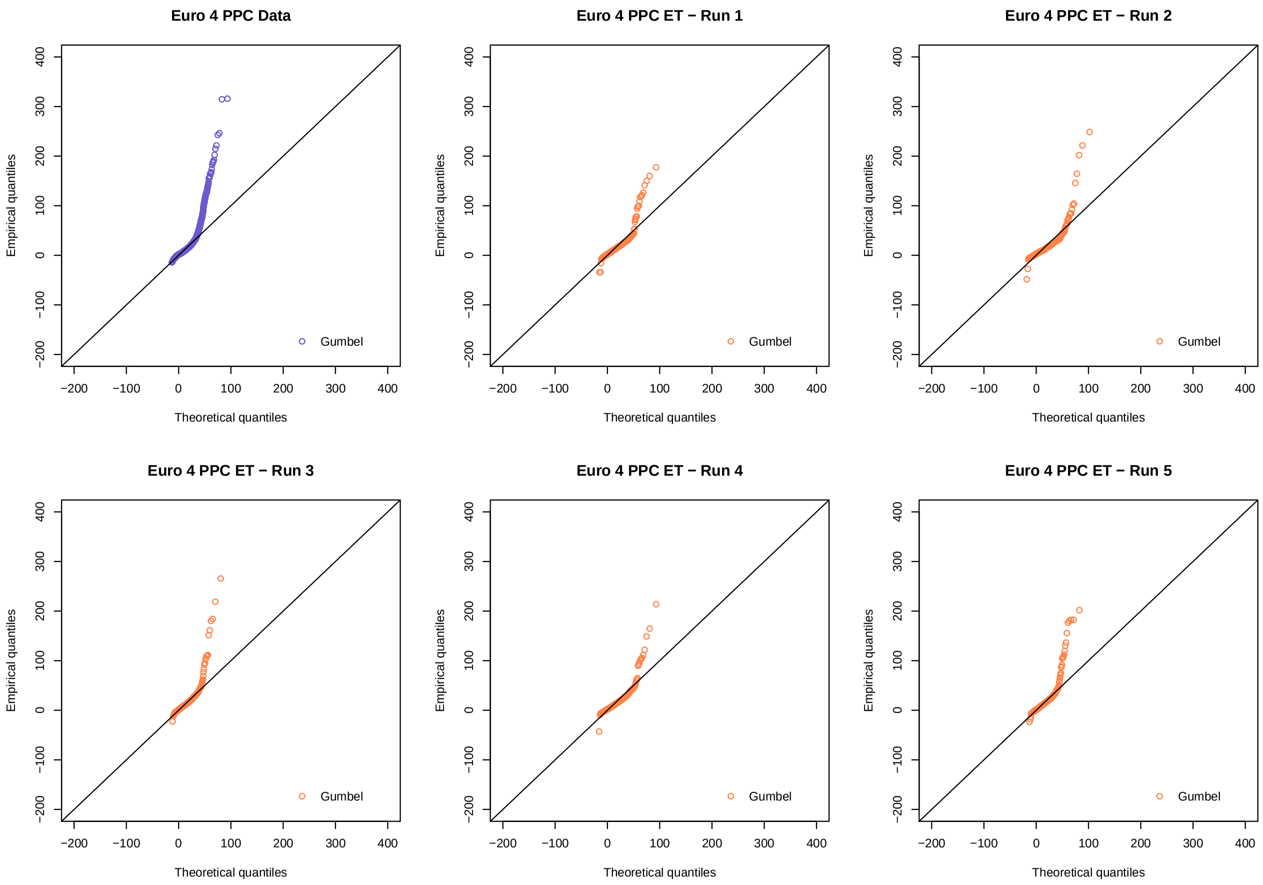

Figure 5.20: QQ Plot of Euro 4 petrol passenger cars modelled using exponential tail

In each plot the bulk of the simulated ratios match well with the observed data. This is largely to be expected as they are simulated using the Gumbel distribution for on-model vehicles. The range and distribution of values are of the most interest in assessing the validity of this model. It is clear that there is a range of maximum values across the presented of the models. This is to be expected as the nature of the methodology not only allows for variation but requires it as multiple models with a range of variations converge to form the solution. The level of variation observed here is slightly concerning however with some models exhibiting especially high variations. For example simulation 3 for Euro 6 diesels is over twice the maximum of the data. In the Euro 6 petrol case simulations 3, 5 and 7 have this feature, 2 and 7 have it in Euro 5 petrol and all of the Euro 4 petrol models have maximum values that are at least double the maximum data value. Whilst these values will be cancelled out by lower than maximum values generated elsewhere in the model these values mean that the exponential tail hypothesis does not accurately model the reality of the fleet behaviour and a better solution may be possible.

The exponential model case better represents the data than the Gumbel distribution alone however there are a number of concerns relating to the highest values generated by it. The hypothesis that the off-model vehicles are caused by failed catalysts, whilst unproved, can be applied using this methodology. The testing of this hypothesis is implemented in a similar way to the exponential tail hypothesis. As with the exponential tail hypothesis the fleet is split into an off and on model fraction and this is determined using the same method as described in Section 5.2.1. In the failed catalyst case the off-model vehicle is designated as Euro 0 for petrol and Euro 5 for diesel. The Euro 0 and 5 vehicles have been observed and their emission ratios have been fitted to their own Gumbel distribution with their own location and scale parameters. The off-model vehicle has its emission ratio assigned in the same way it would have had before but instead of using its classes location and scale parameters the parameters for Euro 0 are used. The resulting fleet is therefore a superposition of Euro N and Euro 0 or 5 vehicles rather than a segregated scenario in the exponential tail case.

Whilst it cannot be ascertained exactly what caused the off-model behaviour a strong candidate for the cause is non-activation of catalyst and the best reference available for this is the measured Euro 0 cars as they have no catalyst. The limiting factor of this methodology is the number of E0 vehicles on the road to be measured. The number of E0 petrol vehicles is 20. Whilst this is enough to fit a distribution function to the error associated with the fitted parameters is higher than if the fleet size had been the size of some other fleet subsections observed. If this method is to be applied in the future the legacy data sets from Aberdeen, Sheffield, Cambridge and other older studies not part of this thesis may need to be combined or Euro 1 vehicles substituted. As with the exponential tail case eleven scenarios were simulated using the Aberdeen data set and observed fleet distribution and are displated in Figures 5.21, 5.22, 5.23 and 5.24. The data is plotted in purple and the simulations are plotted in yellow. The fitted model is overlaid in red.

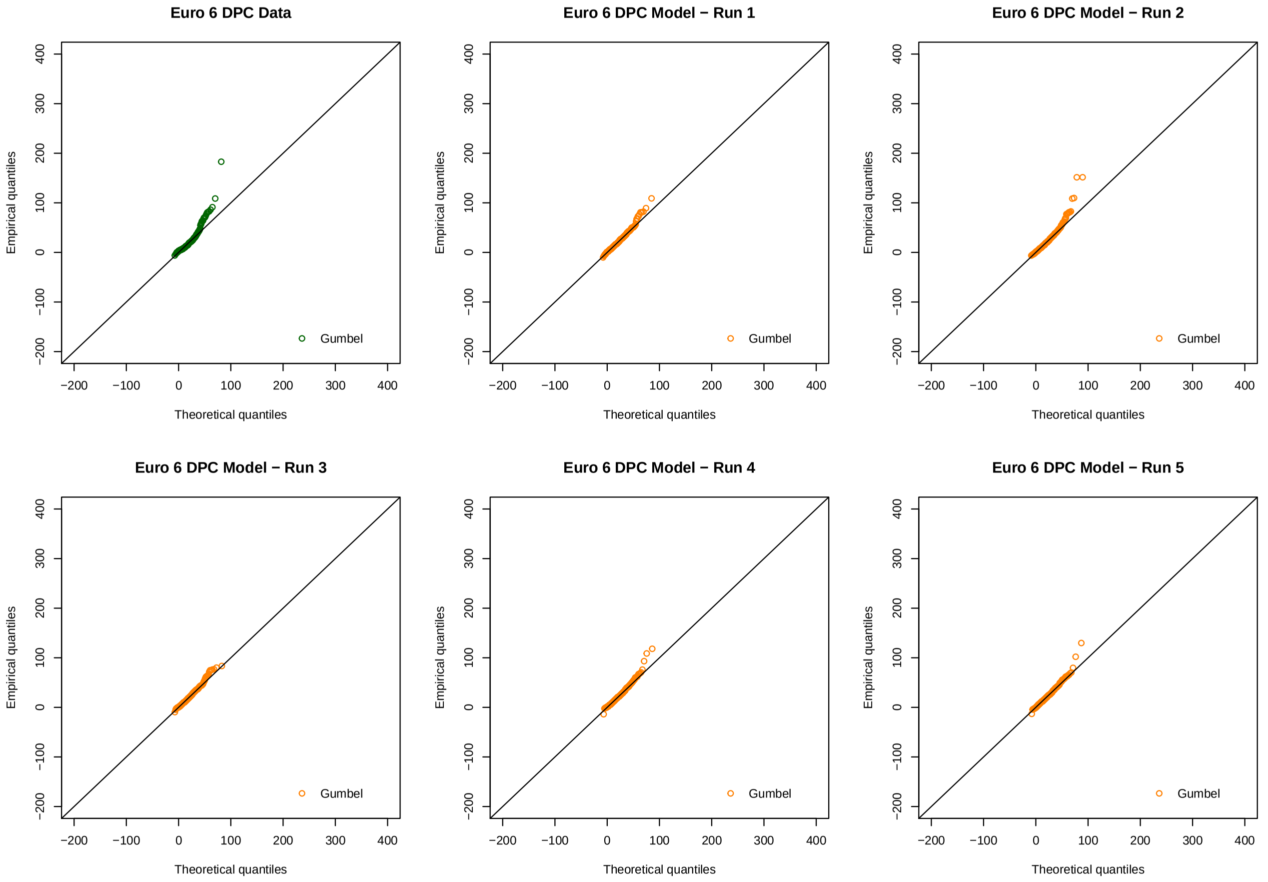

Figure 5.21: QQ Plot of Euro 6 diesel passenger cars modelled using failed catalyst

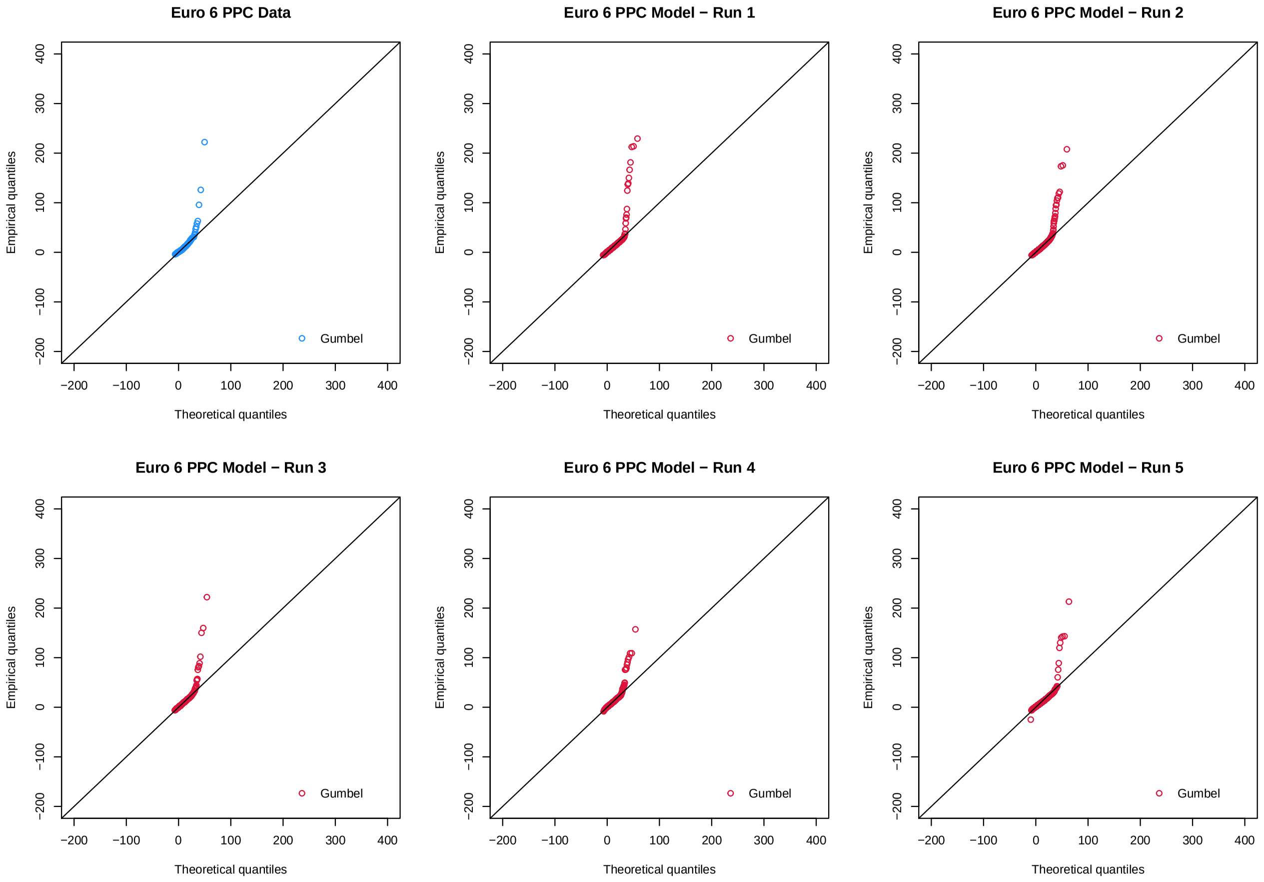

Figure 5.22: QQ Plot of Euro 6 petrol passenger cars modelled using failed catalyst

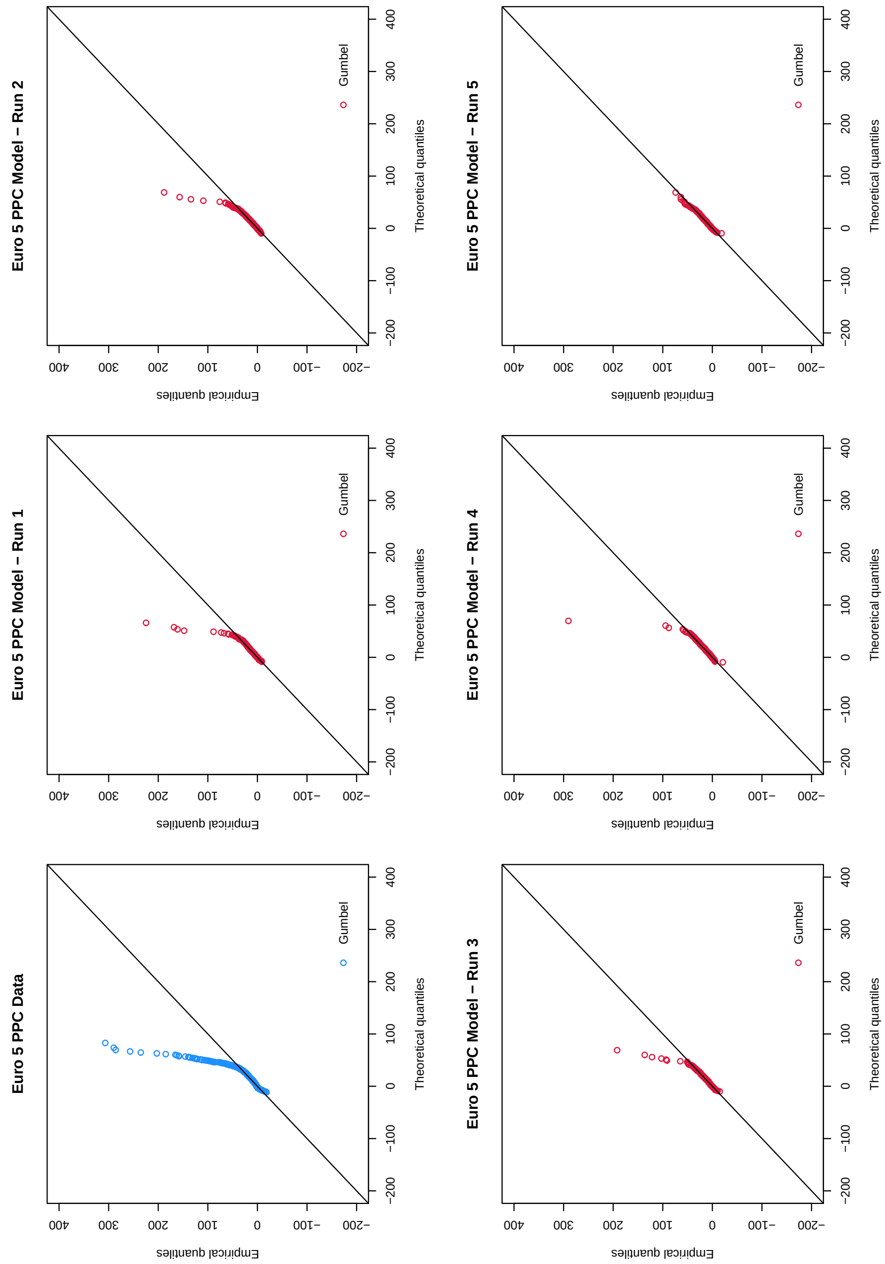

Figure 5.23: QQ Plot of Euro 5 petrol passenger cars modelled using failed catalyst

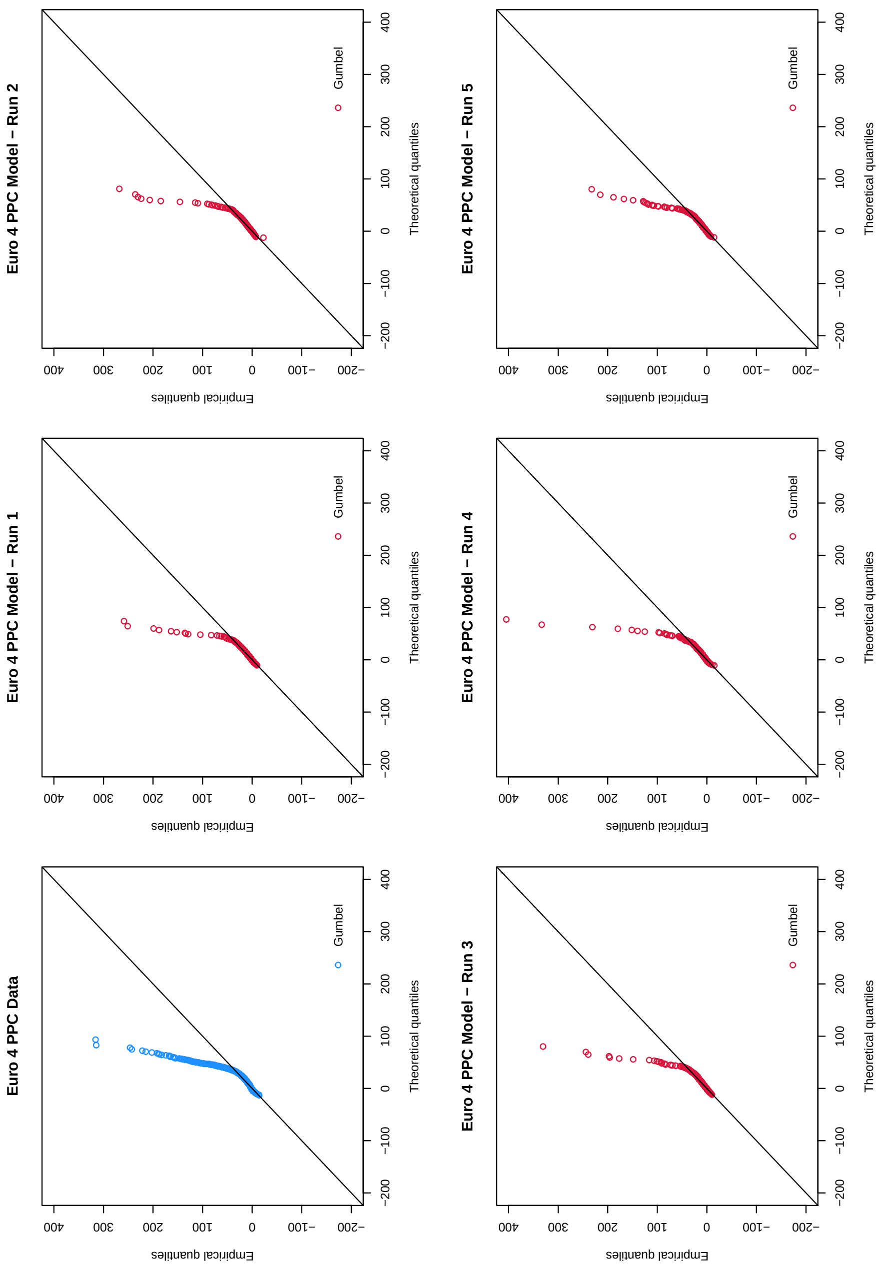

Figure 5.24: QQ Plot of Euro 4 petrol passenger cars modelled using failed catalyst

The failed catalyst model results show a greater level of conformity to the observed data set. This is more obvious as the vehicles get older. Generally any forcing of conformity onto a Monte-Carlo style model is not desirable however in this case the two fleets that form the total are superimposed rather than concatenated and it should not introduce a bias. The solution is based more in reality than simply bolting an additional section onto the fleet distribution. As such modelling the off-model component of the fleet as Euro 0 vehicles appears to produce a satisfying result across the range of results. It remains a plausible hypothesis and a useful tool for modelling however further work must be done to better understand the causes of the off model vehicles. The fact that the use of this approach results in a very good representation of the observed vehicle fleet suggests that for a lot of the off-model vehicles the cause is a failed or otherwise non-operational catalyst system and adds extra weight to the argument to investigate these vehicles further.

To compare the two algorithms the model was run for 35000 cars, similar to the number in the Aberdeen fleet. In one case the exponential tail (ET) algorithm was used and in the other the failed catalyst (FC) algorithm was used. The fleet distribution was the observed distribution in Aberdeen. A comparison between the two algorithms compared to the data for Aberdeen is plotted in Figure 5.25.

Whilst the ET algorithm does not perform terribly compared to the data there are some notable departures from what is observed. The shape of the Q-Q plot distribution is similar to that of the data but as expected from previous analysis the maximum values generated through this method are higher than those observed in the fleet. This result is based only on one model but the number of vehicles sampled means that the full range of the exponential scale will be visible in these plots. The FC algorithm produces results that are largely in agreement with the observed data. This is indirect circumstantial

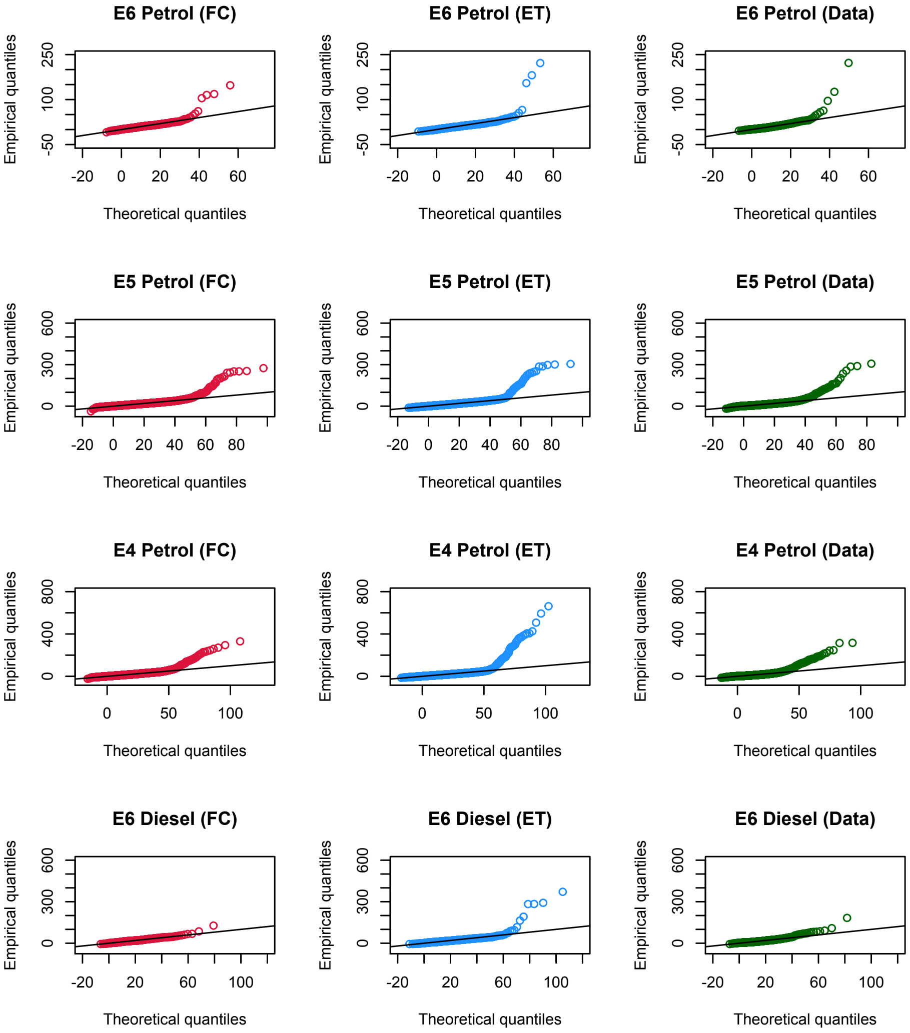

Figure 5.25: Comparison of FC and ET algorithms to observed data

evidence that the cause of the off model observations are at least in part caused by catalysts that are operating closer to that of an E0 vehicle, one that does not have a catalytic converter fitted, backing up the assertion made in Section 5.3. Other influencing factors may still apply however the RSD data does not give any additional levers to control the model. The Euro 6 diesel vehicles do not exhibit any uplift in the high range for the FC model. This is likely to be because of errors introduced due to the small fleet fraction of these vehicles meaning that an off-model Euro 6 diesel vehicle was not generated by the model or the size of the Euro 0 diesel fleet. Given that all the Euro 6 diesel vehicle points are good intersects with the model line it is most likely that it is caused by the non-generation of an off-model Euro 6 diesel vehicle. It is concluded that the failed catalyst hypothesis is the best model to be used for analysis of petrol powered vehicle emissions.

5.5 Conclusions and Discussion

The distribution of vehicle emissions has been assessed using the gamma distribution and the Gumbel distribution. Both distribution functions effectively map the distribution of vehicle emissions for some cases but only the Gumbel distribution is able to describe the vehicle emissions in all cases. The parameters used to describe the Gumbel distribution are more desirable as they are better related to physical parameters of the fleet. The fleet is also shown to be two-typed with a significant departure from the Gumbel distribution in some cases. The number of vehicles that depart from this distribution is as high as 30% for older vehicles but is as low as a couple of percent or non-existent in other cases. It has been shown that the Gumbel distribution is effective across a range of different locations and passenger vehicle types and the difference between these vehicles is further investigated in Sections 7.4 and 7.5. Potential sources of variation that could impact the distribution of vehicle emissions measurements have been investigated and the level of disaggregation required before differences in the distribution has been shown to be at the city level.

The Gumbel distribution does not adequately account for the off-model highly emitting vehicles that make up the top few percentiles of all the petrol vehicle subsets and the latest Euro 6 diesel subsets. It has been observed that the percentile that does not behave in accordance with the Gumbel distribution increases as the age of vehicles increases. It is hypothesised that the ageing of these vehicles is therefore responsible for this departure. The remote sensing device measurement and the associated meta-data does not have any way of assessing the cause of this departure directly however. A number of potential sources of this emissions behaviour are theorised and a number are eliminated. The impact of vehicle specific power, a proxy for engine loading, has been excluded as removing these vehicles from the data set does not remove the departure of the highest emitting vehicles from the Gumbel distribution. It is therefore more likely that the cause of these is related to the physical state of the vehicle catalyst systems.

There are a number of potential causes for the departure that are discussed in the chapter. A catalyst system that is not running at optimal conditions will not be as effective at reducing emissions as those that are. The sub-optimal catalyst running could be caused by a failure to achieve catalyst light-off through a cold start. It is not possible for the RSD to explicitly test for the temperature of the catalyst in a real driving emissions survey however in further work the analysis of cold starts could be achieved through controlled remote sensing surveys designed to investigate this phenomena. An attempt to tie observations of the CO and HC emissions and hence cold start conditions have been attempted but are currently inconclusive. Further work in this area focussing on colder operating conditions may be able to better identify cold-start engines. It has also been shown in studies that vehicles have their emissions controls switched off or reduced at colder temperatures. A relatively basic attempt to test this using remote sensing could be attempted as further work using the ambient temperature measurements taken as part of the RSD measurement however data relating to the vehicles' onboard temperature sensors would be better as that is what the vehicle is using to control the emissions system. Finally the general ageing through poisoning, damage and deactivation of catalyst sites in the converter may be causing these observations. Without the additional information required to back up the cold-start and cold running hypotheses it is taken that these vehicles are running with failed catalysts.

A model has been proposed that uses the mathematical description presented in this chapter to describe the whole fleet emissions. The model describes the fleet as behaving in two characteristic parts defined as on-model and off-model. The on-model vehicles typically account for $\approx 90\%$ of the vehicles of a given subsection and their emissions behave in line with the Gumbel distribution and can be modelled as such. The off-model fraction account for the rest and emit more highly than would be expected from the observed model. Two hypotheses have been proposed and tested to both describe and understand the source of these emitters. The exponential tail and failed catalyst hypotheses have been tested and the failed catalyst hypothesis has been shown as superior.

The success of the failed catalyst model to agree with the observed data displayed in Figure 5.25 is an encouraging result. With any model the less fine tuning that is required the more satisfying it is when the result matches reality. Given that the results match so well with the observed data without the need to adjust additional parameters beyond setting the off-model fraction models used throughout the rest of this thesis we assume that the underlying cause of off-model vehicle behaviour is due to a failed catalyst. Whilst it still remains to be confirmed through external testing using additional measurement methodologies this solution is both plausible and effective. For this methodology to be continued the legacy data observed by the RSD must be used to ensure that a range of Euro 0 vehicles are always available to create distributions from. The Aberdeen set had 20 Euro 0 petrol and 3 Euro 0 diesel vehicles in the whole fleet and as these vehicle are removed from the fleet they will not be able to be used as a direct resource to estimate the failed catalyst rate. As such the vehicles already measured represent the only resource left to estimate failed catalyst models. It may be possible to use Euro 1 vehicles as a surrogate for Euro 0 if in the future not enough Euro 0 vehicles are available to fit a distribution to.

Chapter 6

The Effectiveness of Euro 6 Technology

6.1 Introduction

The introduction of Euro 6 technology has the potential to be a key turning point in the reduction of NO_X emissions from diesel cars. The success or failure of the legislative steps to do so have far reaching implications. As the problems with older generations of legislative steps are becoming clearer the drive to reduce NO_X in real terms has increased. With the WLTP and real driving limit values being proposed for future type approvals of Euro 6b and Euro 6c the current Euro 6 legislation is the last chance for manufacturers to get it right in the current testing paradigm. Euro 6c legislation, that will come into force in September 2017, will keep the same NO_X limit values as previous Euro 6 legislation however the vehicle will have to meet these limits using the WLTP drive cycle and real driving PEMS previously vehicle would only have to pass the limit values using the NEDC (Section 2.4). The real driving PEMS conformity factors are currently set at 2.1 for new cars by 2019 (new models 2017) and decrease to 1.5 by 2021 (new models 2020). The real driving criteria consists of a 90-120 minute drive by a normal driver on urban, rural and highway roads. Remote sensing measurements of current Euro 6 vehicles in real driving environments will be able to both asses the effectiveness of the Euro 6 technology as it is currently applied. Remote sensing measurements of current vehicles will also provide a baseline to compare with future measurements so any effects that changing from an NEDC to a WLTP and PEMS based system has had on real driving emissions.

In local terms the Low Emission Zone (LEZ) in London, the largest in the world, has generally been considered a failure as it has not significantly reduced concentrations of PM_{10} in the city centre and although not a stated aim has failed to reduce the total NO_X compared to non-LEZ levels (Ellison *et al.*, 2013). The new Ultra-Low Emission Zone (ULEZ) due to be enforced from 2020 in London is set to prohibit all diesel vehicles that do not meet the Euro 6 type approval standards from the city centre. It will also prohibit access to petrol vehicles that are do not meet Euro 4 emissions levels. Older vehicles that do not comply with these limit values will be fined £12.50. The success or failure of the Euro 6 technology will impact on the success or failure of the ULEZ to meet it's goal of improving the air quality in the city and bring it into compliance with the EU regulations regarding ambient pollution concentration.

The goals of this chapter are as follows: *i* To analyse the Euro 6 passenger car emissions data collected from Leeds and Aberdeen in particular as the number of Euro 6 vehicles observed in these data sets is higher than those in Sheffield and Cambridge. It will also look at data from Zurich where Euro 6 vehicles have been driven along a motorway at high speed for an extended period of time. After the analysis of these data they will be compared and contrasted with the data relating to Euro 4 and Euro 5 diesel cars to try to assess what, if any, improvement has been made. *ii* To use the new information on emission factors and the NAEI fleet projections (2011 base) to project the total NO_X reduction across the fleet by 2025. *iii* To assess the impact of off-model vehicle contribution to this model.

To achieve these goals the passenger vehicle fleets from Aberdeen, Leeds and Zurich are fitted to a Gumbel distribution as described in Chapter 5.4. The fleet is then recreated virtually using Monte-Carlo methods based on changing fleet composition to estimate future NO_X emissions. These predictions will give information about how effective the new cars are as well as their contribution to the total inventory. It will also provide a baseline do nothing scenario to which other policies can be tested in Section 7.2. The hypothesis presented in Section 5.5, that the off-model vehicles are caused by failed catalysts is also tested by modelling them as Euro 0 vehicles.

6.2 Reduction in Emission Factors for Euro 6 Vehicles

6.2.1 Zurich Measurements

To assess the improvements in emission factors for Euro 6 vehicles compared with Euro 5 and older the fitted location and scale parameters for the NO emission factor in parts per million $[ppm]$ returned from the remote sensing device from the Euro 6 Zurich diesel passenger vehicles were compared to the location and scale parameters for the Euro 5 and Euro 4 diesel vehicle fleets. No vehicle base CO_2 values were available so total emission factors (gkm^{-1}) were not calculated. Whilst less representative of total real world NO_X emission in gkm^{-1} than a fully calculated emission factor this metric still allows a comparison to be made between the three fleet subsections. The results for this analysis are presented in Figure 6.1 and Table 6.1.

A ten-fold reduction in the ratio of $NO : CO_2$ is observed between Euro 6 and Euros 5 and 4 class vehicles. If this order of magnitude

Fleet Section	Location (a [ppm])	Scale (b)
Zurich		
Euro 6 Diesel	41 ± 7	10 ± 6
Euro 5 Diesel	442 ± 4	331 ± 3
Euro 4 Diesel	408 ± 5	293 ± 4

Table 6.1: Quantitative comparison between NO emissions for Euro 6, Euro 5 and Euro 4 diesel vehicles in Zurich

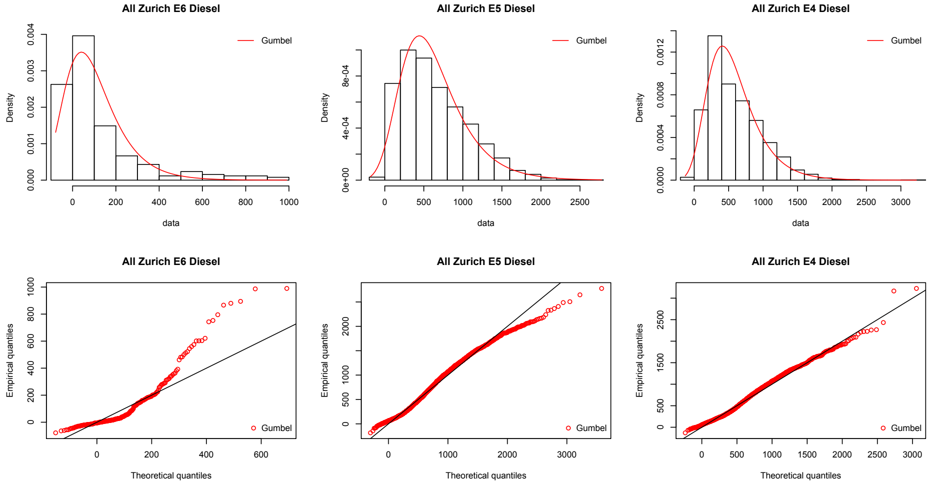

Figure 6.1: Distribution and QQ plots for NO ppm emission for Euro 6, Euro 5 and Euro 4 diesel vehicles in Zurich.

change is to be carried over to the total emission factors in urban environments then the results would show that the Euro 6 technology emits significantly less total NO. The Q-Q plot for Euro 6 suggests that there is evidence of additional super highly emitting vehicles compared to the previous Euro standards however these vehicles are still emitting in the normal range for Euro 5 and Euro 6. The Zurich dataset offers opportunities to observe vehicles after hot running where the vehicles' emissions reduction systems should be operating at their optimum operational temperatures. In this case it appears that the NO emissions have been significantly reduced. This site is not typical of all urban driving and expanding this analysis across a range of different sites is required to further understand the emissions of Euro 6 vehicles.

6.2.2 Urban Driving Environment - Aberdeen and Leeds

The Zurich analysis is extended to the Leeds and Aberdeen data sets. The fitted location and scale parameters of the NO_X emission factors in gkm^{-1} calculated from the Aberdeen $NO : CO_2$ measurements using Equation 3.5 are presented in Table 6.2. The results show that there is a clear reduction in peak total NO_X emission factor and a reduction in the influence of the high emitting tail as well. Whilst the magnitude of the difference is significantly less than observed in the Zurich data set the reduction in NO_X is still significant. It is likely that the cause of this is the environment of vehicle operation. The vehicles measured in the Zurich study have been running up-hill for an extended period of time and this is the ideal operating conditions for emission control systems. The urban environments in Aberdeen and Leeds are less ideal. Figure 6.2 shows the difference in the population density and QQ plots for the fits. There is still a deviation from the theoretical model caused by the most highly emitting vehicles in the Euro 6 fleet however their impact on the population as a whole has

Fleet Section	Location ($a\ [gkm^{-1}]$)	Scale (b)
Aberdeen		
Euro 6 Diesel	0.39 ± 0.02	0.33 ± 0.01
Euro 5 Diesel	0.77 ± 0.008	0.598 ± 0.007
Euro 4 Diesel	0.80 ± 0.01	0.65 ± 0.01
Leeds		
Euro 6 Diesel	0.35 ± 0.06	0.26 ± 0.04
Euro 5 Diesel	0.69 ± 0.02	0.53 ± 0.02
Euro 4 Diesel	0.75 ± 0.02	0.57 ± 0.02

Table 6.2: Quantitative comparison between Euro 5 and Euro 6 diesel vehicles in Aberdeen

decreased from the previous two Euro classes and the number that fall into that range are less than is observed in the Zurich data set.

The analysis was also performed on the smaller sample of vehicles measured in Leeds between June and July 2014 (Section 3.4.3) with the results also presented in Table 6.2 and Figure 6.2. As with the Aberdeen data set the location parameter is approximately half the location parameter fitted to the Euro 5 and Euro 4 diesel data sets. The scale parameter of the Euro 6 fit is less than half that of the Euro 5 and Euro 4 diesel fleets. This represents a significant decrease in total NO_X emitted by these vehicles.

6.2.3 Summary

It has been shown through observations at three different sites in two different countries that the emission of NO from Euro 6 diesel passenger cars has reduced from Euro 5 and Euro 4 levels. It has also been shown that these vehicles are at their best after an extended period of higher loading as observed in Zurich. Urban driving measurements confirm that the Euro 6 emissions are less than those of

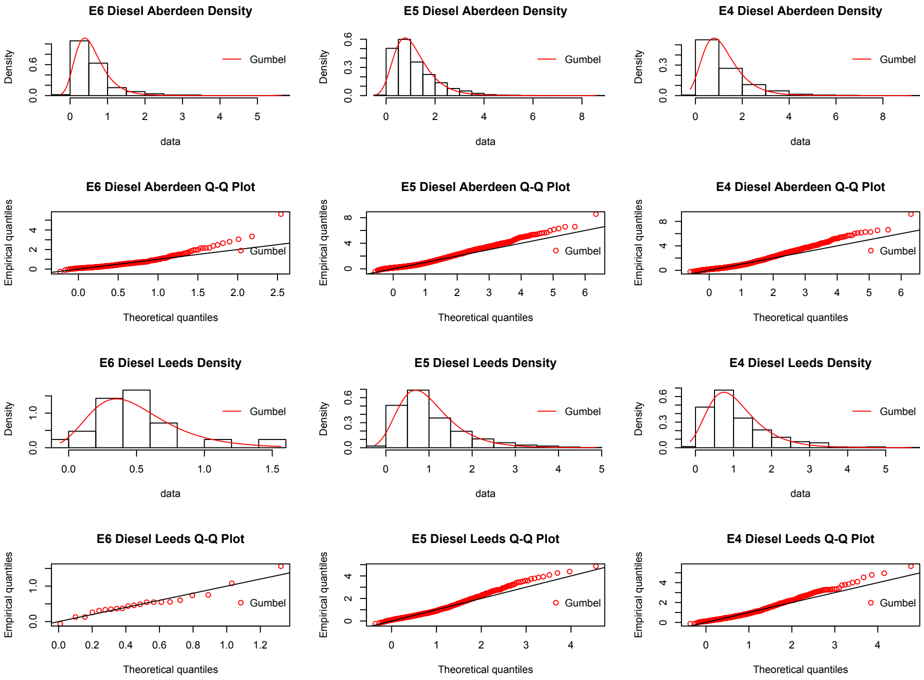

Figure 6.2: Distribution and QQ plots for NO_X emission factors over Euro 6, Euro 5 and Euro 4 diesel vehicles in Aberdeen and Leeds.

Euro 5 and Euro 4 vehicles however the effect is less pronounced in these situations. The impact that these vehicles will have on the total fleet emissions of NO_X will be investigated in Section 6.3.

The number of off-model vehicles observed in Euro 6 diesel powered vehicles is considerably higher than observed in any other diesel powered Euro class subset. The existence of these vehicles has been noted in previous publications (Franco *et al.*, 2014; Yang *et al.*, 2015a,b) and their presence in the on-road fleet appears to have been confirmed by remote sensing measurements. Off-model vehicles have been observed in other data-sets and some hypotheses as to their cause have been proposed in Section 5.4.2. These hypotheses are tested in Section 5.4. These vehicles may be a cause for concern in the future if they are caused by failed or non-operating catalytic converters as they could reduce the effectiveness of legislative steps focused on banning vehicles below Euro 6 for diesel power.

6.3 Modelling The Fleet

It has been shown that relying on the mean and standard deviation to describe the emissions of fleets as a whole is not appropriate. A new mathematical framework for describing the fleet has been proposed using the Gumbel distribution (Section 5.4). Previous work has used the Gamma distribution however for comparisons between fleet subsets the Gumbel distribution is more useful and no less accurate. Using remote sensing data gives us the opportunity to view a considerably larger selection of the fleet than has been available using PEMS or alternative measurements. The broader range of vehicles measured means that distribution functions fitted to the observed fleet are more representative of the whole fleet than otherwise would be achievable. Combining accurate fleet emissions data and the distribution of vehicle Euro classes and fuel types allows for a more robust assessment of the potential changes in emissions when policy is changed. It is hoped that an improvement in predictive ability will allow policy makers to make better decisions about how to implement low emission zones (LEZs) and ultra-low emission zones (ULEZ's) in areas where NO_X concentrations are especially high.

This section models the levels of emission reduction that could be expected in do-minimum case where the fleet is left to naturally evolve using the methodology described in Section 5.4. Two main scenarios are tested. The on-model only scenario where vehicles that are in the off-model section of the distribution are ignored and a two-subset scenario which tests both the exponential tail method and the failed catalyst method for modelling the off-model vehicles. The technical details of assessing the off-model fraction using the failed catalyst method are investigated. The difference in emission factors across these three models is presented at the end of the section.

6.3.1 Vehicle Type Distribution

The fleet projections used in this section are based on he NAEI fleet projections (NAEI, 2012) from a 2011 base, calculated using total vehicle kilometres traveled by the whole fleet have been subsetted into Euro classes 0 through to 6 and further into fuel types petrol and diesel. Given the time constraints of this project the fleet is not further split into hybrid vehicles however this remains a topic for future work. The purpose of this model is to assess how the emissions inventory of the fleet will evolve up to the year 2025 and to provide context for further investigations into the impact of policy changes undertaken in Section 7.2. The fleet splits for the years from 2015 to 2025 are analysed individually to create a year by year projection of NO_X emissions across the network. The distribution of vehicles in Scotland is validated using the on-road fleet distribution observed in the meta-data acquired as part of the remote sensing survey and shows that in Aberdeen there are considerably more diesel passenger

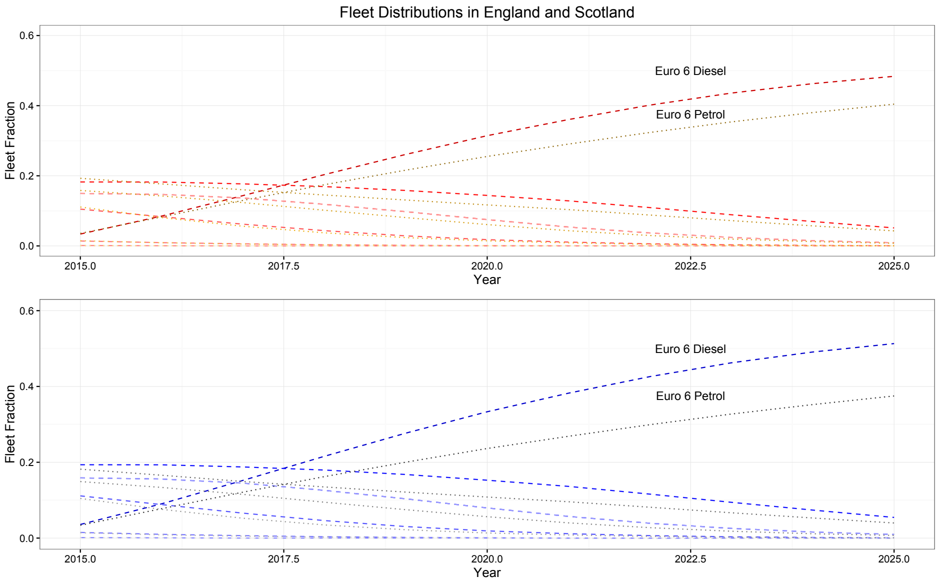

Figure 6.3: Fleet changes in England (red) and Scotland (blue) 2015 – 2025

cars than would be predicted by the NAEI (Figure 7.1). The fraction of each vehicle class is shown in Figure 6.3. The data is colour coded by Euro class going from lightest (E0) to darkest (E6) blue and silver and red and gold for Scotland and England, diesel and petrol respectively.

The analysis shows that the number of older vehicles is representative of the vehicles observed on the road in Aberdeen however the number of Euro 5 diesel vehicles is significantly higher than might be expected and both Euro 3 diesels and Euro 6 diesels and petrols are under represented in the NAEI projections. This is likely to be because of the difference in wealth present in Aberdeen compared to other areas of Scotland. Aberdeen is renowned for its oil industry and the Bridge of Dee observation site was located on a key commuter link on the network. The NAEI fleet projections were for the whole of Scotland which experiences a broader range of economic circumstances. Further influencing the difference between the NAEI predictions and the observed numbers was the time of year. The measurements were taken relatively early and cannot account for new vehicles purchased later in the year. The same projections are available for England, Wales and Northern Ireland. In this experiment the emissions inventories for both England and Scotland are estimated and compared. For the purpose of projecting future emissions values and with the lack of a more bespoke value for Aberdeen the projected fleet distribution estimated by the NAEI are considered acceptable.

6.3.2 On Model Vehicles

It has been shown in Chapter 5 that both the Gumbel distribution and the gamma distribution realistically represents a large section of the vehicle fleet with the Gumbel distribution proving to be a more useable framework with fit parameters that are better related to real features. Various tests and limitations have been performed and analysed using the Gumbel distribution to assess its validity in describing the emission factors of passenger cars driving in an urban environment. The Gumbel distribution has been shown to be successful at describing all but the most highly emitting vehicles. It is hypothesised that these vehicles possess some characteristic that is atypical of the fleet as a whole but nonetheless needs to be recognised. Modelling the fleet using the Gumbel distribution along with the caveat of the off model high emitters is therefore a valid mathematical model for the passenger car fleet as a whole. The impact of Vehicle Specific Power (Section 5.3.2) and site selection (Section 5.3.3) have been shown not to influence the distribution and presence of the atypical off model super highly emitting vehicles.

The emission factors are calculated from the Aberdeen data set (Section 3.4.4) as it represents the most up to date data that is available and also contains enough Euro 6 vehicles for the model to accurately fit a distribution to the data. To assess the on-model fleet is broken down into subsections by Euro standard and fuel type. Each of these subsections have a Gumbel distribution function fitted to them and the model parameters are stored in a matrix referred to as the Location-Scale matrix (LS matrix) which is accessed by the model later. The fleet is then resampled using a random number *Monte-Carlo* method to create a virtual fleet made of virtual cars. Random assignment based on the pre-determined fleet split assign a Euro class and fuel type of the vehicle and this process is repeated until the number of virtual vehicles created is the same size as the desired virtual test fleet. Each new vehicle is assigned an emission ratio randomly from a Gumbel distribution that with matching location and scale parameters from the LS matrix entry for its fuel type and Euro class. The total $NO_X : CO_2$ ratio is calculated using the fractional NO_2 values measured in Carslaw & Rhys-Tyler (2013) and validated in Chapter 4. An externally estimated CO_2 emission factor is then combined with the emission ratio to calculate the NO_X emitted by each vehicle. Vehicle CO_2 emission factors are extrapolated across the network using the vehicle tracking and PHEM method described in Section 2.5.3, Tate (2016) and Wyatt *et al.* (2014) as this step will further improve the efficacy of the model by relating it to the network it was measured in rather than using laboratory derived CO_2 factors. The full fleet and its total NO_X emission factors have now been created. The aggregated NO_X per vehicle can then be assigned by calculating the mean NO_X emission factor. As with all Monte-Carlo type simulations the process is repeated a number of times until a convergence is reached. For the purpose of the future projections section the number of virtual vehicles is 15×10^3 vehicles ran over 10 simulations per year per location. Data from Scotland and England are presented resulting in 200 simulations being ran per case and 30×10^5 total virtual vehicles being assessed. The approximate time per simulation is $30s$ using a commercially available laptop fitted Intel i7 2.2GHz processor running the R program in its standalone form.

6.3.3 Off Model Vehicles

For the vast majority, often greater than 98%, of vehicles the Gumbel distribution realistically describes the behaviour of a fleet subset as a whole however in most subsets of the vehicle fleet there are vehicles that have emissions which do not fit the Gumbel distribution. For the purposes of modelling these vehicles the cause of this off-model behaviour taken to be the failed catalyst hypothesis described in Section 5.4.2. To accurately describe each subset and it's contribution to the total emissions these super highly emitting vehicles must be accounted for. In this section the hypothesis asserted in Section 5.5, that the cause of these off-model vehicles is failed catalysts, will be tested by replacing a fraction of the vehicles in each fleet subset with either a Euro 0 petrol vehicle or a Euro 5 diesel. For petrol vehicle these are vehicles that are not subject to any limit value legislation simulate a failed or otherwise non-operational catalyst in the model. For diesel vehicles the Euro 5 legislation is chosen in line with the NAEI method for estimating a failed catalyst.

For this scenario to be tested at the point of calibration of the model the user must identify the percentile where the vehicles stop behaving in line with the model and start exhibiting characteristics of super highly emitting vehicles. The method for setting the off model quantile is described in Section 5.2. The Q-Q plots for all the passenger vehicle subsets in Aberdeen are shown in Figure 6.4. The Gumbel distribution function is shown as a black line. The on and off model sub-fleets are clearly distinguishable. On model vehicles track the black line and off model vehicles do not. Estimating the fraction of vehicles off line is achieved by cutting the data at various quantiles and trying to get the best match of the remaining data to the model line. Figure 6.5 shows the Euro 6 diesel fleet for Aberdeen with various top percentiles removed. For example the 99% plot is the fleet

Fuel	Euro 0	E1	E2	E3	E4	E5	E6
Off Model Quantiles							
Petrol	1.0	1.0	0.70	0.902	0.967	0.986	0.98
Diesel	1.0	1.0	1.0	1.0	1.0	1.0	0.95

Table 6.3: Off model quantiles for vehicle classes modelled in Aberdeen

with the top 1% removed. The cut percentile is optimal when the predicted and theoretical quantiles best align on the presented 1:1 line. In this example a case can be made for setting the percentile where the theoretical quantiles and the empirical quantiles best match up is at either 95% or 96%. To model this fraction the percentage point represented by ξ_{fleet} is expressed in the decimal form so $\xi_{95\%} = 0.95$. After the limiting point has been determined a second Monte-Carlo random number is generated between $0 - 1$. If the number is less than the ξ_{fleet} then it is designated off-model and it's emission ratio is resampled from the Gumbel distribution representative of it's subset. If it is greater than ξ_{fleet} then the vehicle is designated as off-model and either the failed catalyst algorithm is applied. Fleet subsections such as the Euro 5 diesel fleet that conform to the model throughout are modelled using their Gumbel distribution in their entirety. The fractional estimations of off-model vehicles for the Aberdeen fleet are shown in Table 6.3.

6.4 Projecting Future NO_X Emission Factors

The following section shows the results of the fleet emissions projections based on the model described in Section 6.3. Projections from models using on-model vehicles only are compared and contrasted with models incorporating the failed catalyst off-model vehicle scenario.

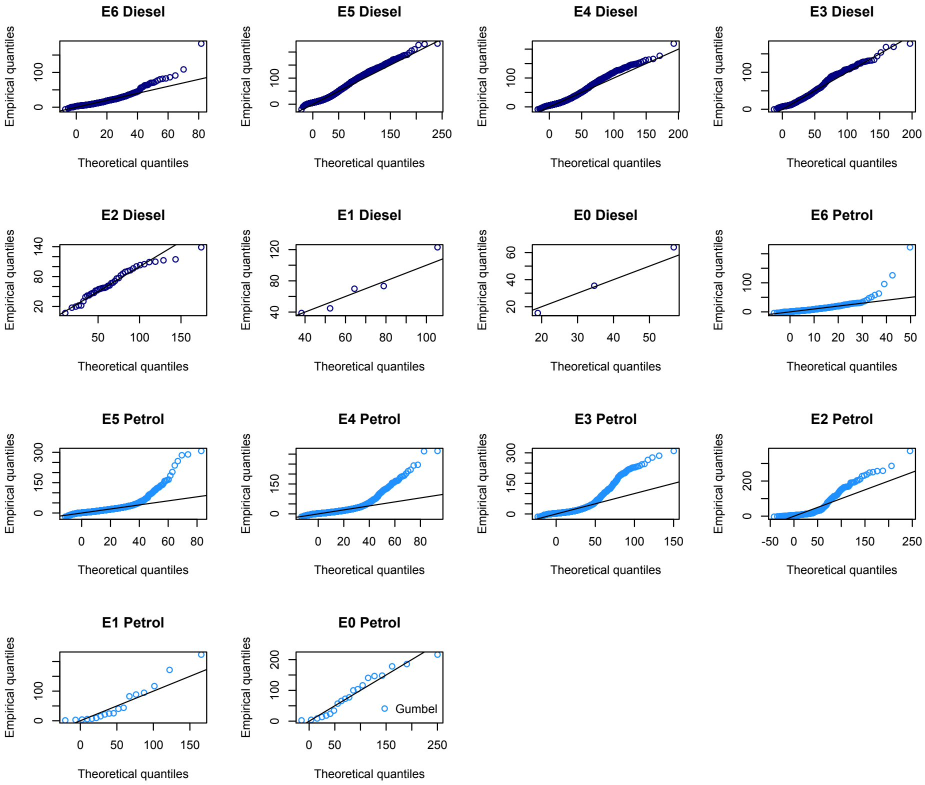

Figure 6.4: Q-Q plot by percentile of the E6 Diesel Aberdeen fleet

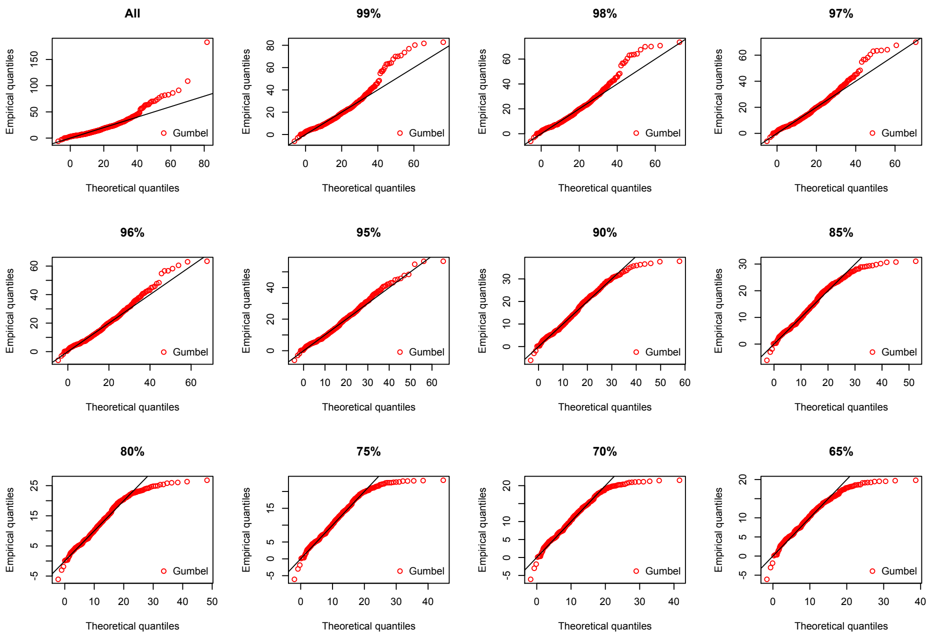

Figure 6.5: Q-Q plot by percentile of the E6 Diesel Aberdeen fleet

The two modelling procedures are completed for both the English and Scottish fleets.

6.4.1 On Model Fleet Projections

To assess the projection in NO_X emissions from the on-model fleet only the model was run for England and Scotland as described in Section 6.3. No account was taken for vehicle ageing. It was assumed that a Euro 6 diesel vehicle in 2015 will behave the same way as a Euro 6 diesel vehicle in 2025. Given that in the Euro 5 and Euro 6 diesel data sets there is no evidence of significant off-model behaviour and asserting that this off model behaviour is caused by ageing processes validates the assumption that no change to the emission ratio distribution due to ageing however future RSD studies should take note of any additional changes in on-model behaviour. The contribution of off-model vehicles is suppressed in the first section. The results for Scotland and England are presented in Tables 6.4 and 6.5. Figure 6.6

shows the same data displayed graphically with England and Scotland shown in red and blue respectively. The limit values for Euro 3 diesel to Euro 6 diesel passenger cars are shown as an indication. A smoothed fit for the average value across the 10 runs is show as a grey dashed line with a 95% confidence interval indicated as a coloured range. The mean values are shown as darker point markers.

Run	2015	2016	2017	2018	2019	2020	2021	2022	2023	2024	2025
1	0.645	0.616	0.592	0.557	0.547	0.516	0.499	0.477	0.456	0.449	0.436
2	0.638	0.614	0.580	0.561	0.535	0.524	0.496	0.479	0.457	0.446	0.437
3	0.646	0.620	0.587	0.559	0.530	0.509	0.501	0.480	0.463	0.445	0.431
4	0.644	0.606	0.592	0.560	0.530	0.514	0.494	0.488	0.460	0.448	0.432
5	0.640	0.617	0.577	0.559	0.539	0.521	0.494	0.476	0.466	0.446	0.436
6	0.648	0.612	0.574	0.562	0.543	0.518	0.491	0.476	0.462	0.450	0.426
7	0.646	0.609	0.583	0.556	0.534	0.521	0.501	0.478	0.465	0.444	0.432
8	0.642	0.616	0.583	0.565	0.537	0.518	0.500	0.476	0.466	0.437	0.435
9	0.645	0.611	0.582	0.567	0.538	0.520	0.492	0.478	0.460	0.447	0.433
10	0.632	0.606	0.578	0.551	0.534	0.512	0.502	0.476	0.460	0.449	0.434
Mean	0.643	0.613	0.583	0.560	0.537	0.517	0.497	0.478	0.462	0.446	0.433
Change	1.0	0.953	0.907	0.871	0.835	0.804	0.773	0.743	0.719	0.693	0.673

Table 6.4: Average NO_X emission in gkm^{-1} for English fleet distribution

Run	2015	2016	2017	2018	2019	2020	2021	2022	2023	2024	2025
1	0.670	0.630	0.601	0.588	0.557	0.525	0.517	0.497	0.477	0.470	0.448
2	0.655	0.638	0.616	0.574	0.551	0.533	0.521	0.496	0.482	0.466	0.448
3	0.657	0.633	0.599	0.578	0.552	0.537	0.525	0.503	0.477	0.462	0.453
4	0.665	0.632	0.608	0.571	0.561	0.536	0.514	0.496	0.482	0.463	0.447
5	0.662	0.639	0.601	0.578	0.557	0.537	0.517	0.493	0.485	0.468	0.443
6	0.660	0.638	0.612	0.590	0.550	0.537	0.507	0.488	0.487	0.465	0.446
7	0.666	0.633	0.606	0.584	0.557	0.536	0.517	0.493	0.483	0.464	0.443
8	0.665	0.643	0.604	0.581	0.547	0.536	0.513	0.504	0.476	0.460	0.447
9	0.662	0.630	0.602	0.580	0.555	0.531	0.513	0.500	0.479	0.463	0.451
10	0.660	0.632	0.604	0.580	0.558	0.531	0.513	0.501	0.476	0.461	0.448
Mean	0.662	0.635	0.605	0.580	0.556	0.534	0.516	0.497	0.480	0.464	0.447
Change	1.0	0.959	0.914	0.876	0.840	0.807	0.779	0.751	0.725	0.701	0.675

Table 6.5: Average NO_X emission in gkm^{-1} for Scottish fleet distribution

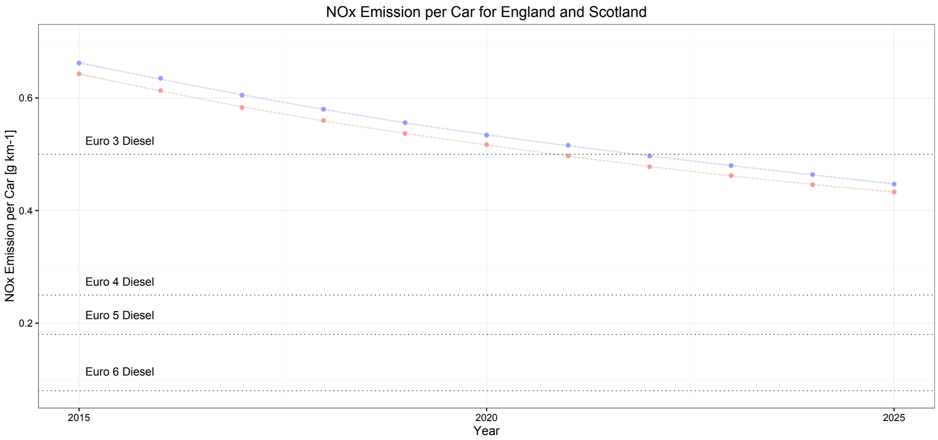

Figure 6.6: Fleet changes in England and Scotland 2015 – 2025 with emissions standards annotated

The results of this analysis show that the total NO_X emitted per vehicle by 2025 has barely fallen below that which would be expected from a Euro 3 diesel passenger car over the NEDC however this decrease does correspond to a figure reduction to 66.4% and 67.2% of 2015 values for England and Scotland respectively. The point at which the decrease in NO_X is comparable to a Euro 3 diesel vehicle is 2021. The observed decrease is due to the number of Euro 6 diesel vehicles that have been added to the fleet at the expense of older more polluting vehicles however despite the improvements seen in the real world effectiveness of Euro 6 emissions standards in reducing the real driving emissions of diesel vehicles there is still a disconnect between the real driving environment and the emissions factors measured in laboratories. The impact of Euro 6c legislation requiring both WLTP drive cycle and PEMS testing to be performed for type approval (Section 2.4) remains to be seen. No Euro 6c vehicles were observed so real world emission factors cannot be calculated or used for forecasting. The results also show that there is less of a reduction in the Scottish fleet than the English fleet. This is likely due to the higher number of diesel vehicles predicted to make up the vehicle fleet by 2025 in Scotland.

6.4.2 Influence of Off-Model Vehicles On Projected Emission Factors

Section 5.4.2 presents the scenario for calculating the influence of off-model vehicles on the total fleet NO_X emission. This subsection includes these vehicles in the calculation of the aggregated emission factor and compares the difference between the on-model only and failed catalyst scenarios.

The models were run with the addition of the algorithm for the failed catalyst algorithm described in Section 5.4.2. The aggregated NO_X emission per vehicle for each run, the mean, the change over time and change relative to the on-model only scenarios are presented in Tables 6.6 and 6.7 for England and Scotland respectively. All the data collected in this Section is plotted in Figure 6.7 with the data from the failed catalyst presented as gold points and navy points for England and Scotland respectively. A LOESS fit smoothed average is plotted with a 95% confidence interval for all cases and is represented by the shaded area. For the failed catalyst scenario the 95% confidence interval is much smaller than for the other two cases and is barely visible in either plot.

Run	2015	2016	2017	2018	2019	2020	2021	2022	2023	2024	2025
R1	0.670	0.631	0.618	0.590	0.562	0.537	0.524	0.509	0.485	0.484	0.460
R2	0.661	0.638	0.613	0.580	0.558	0.537	0.522	0.509	0.493	0.473	0.458
R3	0.662	0.645	0.610	0.590	0.573	0.536	0.526	0.504	0.491	0.475	0.463
R4	0.667	0.629	0.603	0.569	0.558	0.546	0.526	0.509	0.500	0.477	0.465
R5	0.669	0.638	0.601	0.589	0.565	0.539	0.521	0.501	0.491	0.472	0.458
R6	0.672	0.643	0.604	0.587	0.549	0.552	0.528	0.511	0.490	0.476	0.464
R7	0.668	0.642	0.607	0.594	0.560	0.538	0.522	0.504	0.488	0.478	0.460
R8	0.667	0.641	0.612	0.594	0.561	0.535	0.529	0.509	0.497	0.471	0.461
R9	0.664	0.639	0.605	0.582	0.566	0.535	0.524	0.503	0.494	0.474	0.459
R10	0.676	0.644	0.606	0.581	0.559	0.536	0.519	0.508	0.487	0.473	0.460
Mean	0.668	0.639	0.608	0.586	0.561	0.539	0.524	0.507	0.492	0.475	0.461
Change	1.000	0.957	0.910	0.877	0.840	0.807	0.784	0.759	0.737	0.711	0.690
On-model Fraction	1.039	1.042	1.043	1.046	1.045	1.043	1.054	1.061	1.065	1.065	1.065

Table 6.6: Average NO_X emission in gkm^{-1} for English fleet distribution including off-model failed catalyst vehicles

Run	2015	2016	2017	2018	2019	2020	2021	2022	2023	2024	2025
R1	0.690	0.661	0.625	0.604	0.577	0.557	0.537	0.520	0.503	0.492	0.476
R2	0.686	0.660	0.632	0.617	0.583	0.561	0.550	0.529	0.510	0.498	0.477
R3	0.683	0.662	0.632	0.602	0.580	0.561	0.545	0.532	0.512	0.494	0.482
R4	0.694	0.666	0.637	0.600	0.585	0.558	0.540	0.526	0.510	0.493	0.482
R5	0.696	0.655	0.631	0.602	0.581	0.565	0.544	0.525	0.503	0.493	0.485
R6	0.690	0.659	0.628	0.602	0.582	0.557	0.543	0.520	0.512	0.488	0.478
R7	0.693	0.671	0.639	0.604	0.578	0.573	0.542	0.517	0.509	0.486	0.471
R8	0.685	0.660	0.622	0.596	0.573	0.555	0.539	0.528	0.508	0.494	0.481
R9	0.694	0.653	0.629	0.603	0.582	0.563	0.551	0.521	0.505	0.494	0.475
R10	0.688	0.651	0.626	0.606	0.588	0.561	0.541	0.532	0.503	0.495	0.477
Mean	0.690	0.660	0.630	0.604	0.581	0.561	0.543	0.525	0.507	0.493	0.478
Change	1.000	0.959	0.926	0.891	0.856	0.818	0.785	0.753	0.728	0.706	0.684
On-model Fraction	1.042	1.039	1.041	1.041	1.045	1.051	1.052	1.056	1.056	1.063	1.069

Table 6.7: Average NO_X emission in gkm^{-1} for Scottish fleet distribution including off-model failed catalyst vehicles

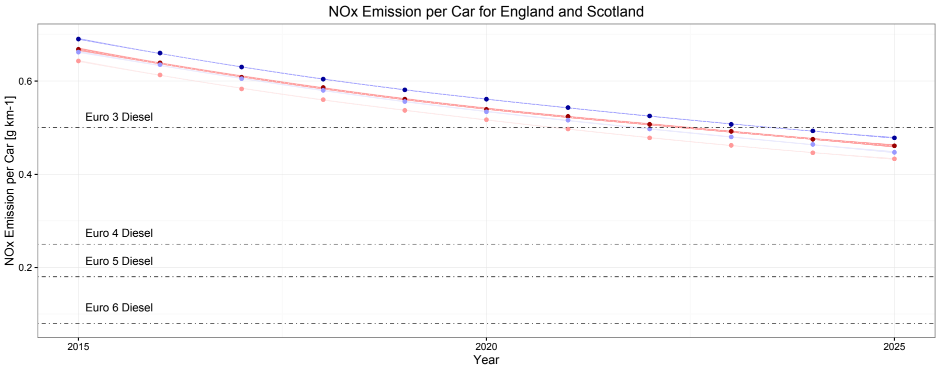

Figure 6.7: Fleet changes in Scotland and England 2015 – 2025 including off-model failed catalyst vehicles

The influence by the off-model fraction of the fleet is non-trivial. In the failed catalyst scenario the extra emissions caused by the off-model vehicles still represents between 4% and 6% of the total fleet inventory and this value is diverging as newer vehicles are introduced to the fleet.

6.5 Conclusions

This chapter set out to answer how effective the introduction of Euro 6 legislation has been in reducing vehicle emissions compared to previous legislative steps. This has been achieved by comparing the current Euro 6 vehicles on the road in Zurich, Leeds and Aberdeen as well as projecting forward to 2025 and assessing the impact that the move towards a higher fraction of the fleet being Euro 6 would have on the total fleet emissions expressed as the aggregated individual vehicle emission factor.

In urban environments the vast majority ($\approx 95\%$) of Euro 6 diesel vehicles are well described by the Gumbel distribution and the location and scale parameters have been used to characterise the fleet as described in Section 5.4. This analysis of the Euro 6 diesel vehicles in situ compared to older vehicles using the same driving environment has shown that the introduction of Euro 6 legislation has resulted in a reduction in peak NO_X emissions by a factor of ≈ 2 in urban driving and a reduction in the size of the on-model tail as well. The observations of Euro 6 vehicles from the Zurich data set show a ten-fold reduction in peak emission values. The number of off model diesel cars observed in the Zurich data set is higher than seen in urban environments however. It is currently unclear what is causing this effect but it is not observed in Euro 5 and Euro 4 diesel vehicles as the Euro 4 and Euro 5 diesel vehicles are largely on-model.

Compared to the Euro 4 and Euro 5 diesel legislative steps where only a small difference was observed the Euro 6 diesel legislation appears to have been more successful. A clear observable difference can be seen between the Euro 5 vehicles and the new Euro 6 vehicles. The two-fold decrease in emission factor is still considerably higher than the legislated limit value of $0.08gkm^{-1}$ that is the type approval value or the conformity factor of 2.1 currently proposed for Euro 6c real driving emissions tests. The stricter Euro 6b and Euro 6c legislation that will be used from 2017 could not be tested however it is hoped that the use of the WLTP drive cycle and the RDE testing using PEMS will further reduce the emissions factors. Given that a clear reduction in NO_X has been observed from Euro 5 to Euro 6 it is hypothesised that any further decreases in NO_X emissions from Euro 6b and Euro 6c would be observable using the remote sensing device data in this way when these new vehicles are introduced to the fleet then any further decrease in their emissions will be observable. Given the step change in type approval process that will occur when these vehicles are introduced the effectiveness of the new system represents an important area for future work.

Projecting the total fleet emission inventory using a Monte-Carlo modelling process has been attempted with the failed catalyst solution to the off-model problem being presented and implemented. The process has been applied to both the English fleet and the Scottish fleet as they have a slightly different distribution of vehicles. Two scenarios are considered. Firstly a fleet of strictly on-model vehicles, representative of the vast majority of vehicles observed. For a fleet of strictly on-model vehicles the projections presented suggest that over the ten year period from 2015 to 2025 the aggregated single vehicle emission factor decreases by a factor of 0.673 and 0.675 in England and Scotland respectively with a total change in gkm^{-1} emission factors of 0.210 and 0.215 for England and Scotland respectively. This is the approximate equivalent of taking a Euro 4 diesel vehicle (limit value $0.25gkm^{-1}$) off the road for every vehicle currently on the road. The aggregated emission factor in 2025 is $0.433gkm^{-1}$ and $0.447gkm^{-1}$ per vehicle in England and Scotland respectively and this is slightly less than two Euro 4 diesel vehicles on the road for every vehicle on the road.

The second scenario which attempts to simulate the effect of failed catalysts in the model has been completed. The vehicles with failed catalysts contribute between 4% and 6% of the total emission inventory between 2015 and 2025. The divergence from the on-model result in later years suggests that the off model contribution by Euro 6 petrol and diesel vehicles is greater than the off-model contribution of older vehicles which is concerning if the main influencing factor causing these off-model results is ageing as hypothesised in Section 5.2. The cause of the off-model vehicles in such a young subsection of the fleet is unknown however it may be linked to the stricter new regulations requiring increasingly more complex emissions control systems which in turn require the window of operation to be finely tuned to those found in the test procedure for the system to pass.

Whilst the forecasts of the vehicle emissions inventory show a downward trend the total emissions inventory is not decreasing at a great enough rate to make a significant difference before 2025 given the fleet projections used in this model. Even with the high percentage of Euro 6 vehicles expected by 2025 the total fleet NO_X emission will not reduce by much more than 30% of the 2015 values if the current Euro 6 technology represents the best practice at removing it from exhaust gasses, which it does not. This result shows that Euro 6c is required to make a noteworthy difference to total fleet emissions. With the introduction of Euro 6b and Euro 6c the test procedure becomes more representative of real world driving than the current one. At the time of writing no vehicles that meet this criteria have been observed on the road and are not likely to be seen before 2017 and as such the influence that these steps will have has not and cannot be factored into this projection. Given the apparent failure of the current Euro 6 technology to significantly reduce the NO_X emissions the emissions factors that Euro 6b and Euro 6c vehicles are able to achieve will be a key factor in reducing the total NO_X emitted by the fleet from 2017 and beyond. Measuring a real driving environment emission factor using remote sensing to compare the RDE factor to the legislated limit values will be very useful in creating a better projection of the total fleet emission and assessing what if any additional measures need to be implemented to further reduce urban NO_X concentrations.

Chapter 7

Application to Real World Problems

7.1 Introduction

A number of real world problems can be readily investigated using the remote sensing data collected as part of this thesis as outlined in Section 1.3. This chapter seeks to explore these problems and to provide answers that are useful for policy makers and manufacturers equally. This chapter will focus on questions that relate to small subsets of the fleet seeking to answer how effective the introduction of Ultra-Low Emission Zones (ULEZs) in Aberdeen and London would be compared to do nothing scenario estimates calculated in Section 6.4 (Section 7.2), the emissions of vans and other light commercial vehicles and the uptake rate of these vehicles to market (Section 7.3). The emissions of taxis compared to regular privately owned passenger cars is investigated (Section 7.4). An assessment of the Volkswagen Group emissions scandal is also presented using the remote sensing data and will examine vehicles known to be fitted with defeat devices and compare them to similar vehicles that have not been involved in a defeat device scandal at the time of writing. The section relating to light commercial vehicles was presented at the Transport and Air Pollution conference (TAP) 2016. The section relating to taxis was presented at TAP 2014. Data relating to the VWG scandal was used as part of an article by The Mail on Sunday (MoS, 2015).

7.2 Testing The Impact of an Ultra-Low Emission Zone

The low emission zone (LEZ) in London has been unsuccessful in reducing any ambient NO_X compared to sites outside the LEZ (Ellison *et al.*, 2013). Restricting peoples' access to any location or forcing them to change their vehicle can be a source of conflict it is therefore desirable to be able to show that an intervention will not only be successful but that the introduction of an ULEZ is an effective, acceptable and affordable way to reduce emissions beyond that which could be achieved by the natural progression of the fleet. The model described in Section 6.3 can be used to test various scenarios relating to ULEZs. This section examines the fleet in Aberdeen and tests two fleet change scenarios adopted directly after the introduction of an ULEZ. An extreme high spend scenario and an extreme low spend scenario detailed in Section 7.2.1. These two cases will then be compared against the fleet projections for Scotland created using the same model but with fleets changing as projected by NAEI (2012). The base case, as assumed and documented in the NAEI fleet distributions, will act as a control or a do nothing to test whether or not similar reductions would be observed if the fleet was left to evolve naturally and therefore whether or not the introduction of an ULEZ is necessary to achieve the same level of reduction.

7.2.1 Vehicle Fleet Distributions

The fleet distribution for Aberdeen in 2015 is known through direct observation. The distribution of the fleets for 2016 - 2025 in Section 6.3.1 are based on projections. Figure 7.1 shows the difference between the observed Aberdeen fleet and the projected average 2015 Scottish fleet. There are a significantly higher number of Euro 5 diesels present in Aberdeen than in the Scottish fleet. As discussed in Section 6.3.1 the smaller number of Euro 6 vehicles in the Aberdeen fleet is likely to be because the survey was conducted towards the start of the year in April and many people would not have upgraded their vehicles yet. The high number of younger Euro 5 diesels compared to the Euro 3 vehicles is likely to be because of the additional wealth in Aberdeen, a city with an oil industry, compared to the rest of Scotland. It is therefore difficult to assert that the Scottish projected fleets would be wholly representative of the Aberdeen fleet by 2025 however without data about how the fleet will evolve this solution remains the best attempt. Accurately modelling the choices that go into people changing their vehicles is beyond the scope of this thesis however when a better fleet projection is available the projections for the Aberdeen fleet should be updated to reflect new information. The Aberdeen 2015 fleet is therefore used as the base scenario and any modifications to reflect an ULEZ are compared to this initial time (t_0) scenario. The following rules apply to both scenarios:

- The fleet is comprised of cars
- The initial fleet distribution is that which was measured in the Aberdeen 2015 study
- No contribution is made to the total NO_X by LCVs and HCVs
- The start date for the ULEZ is 1 May 2015
- The changeover period is assumed to be a step change
- The ULEZ follows the vehicle criteria in the London ULEZ (Section 2.4.5)
- The conformity rate is 100%

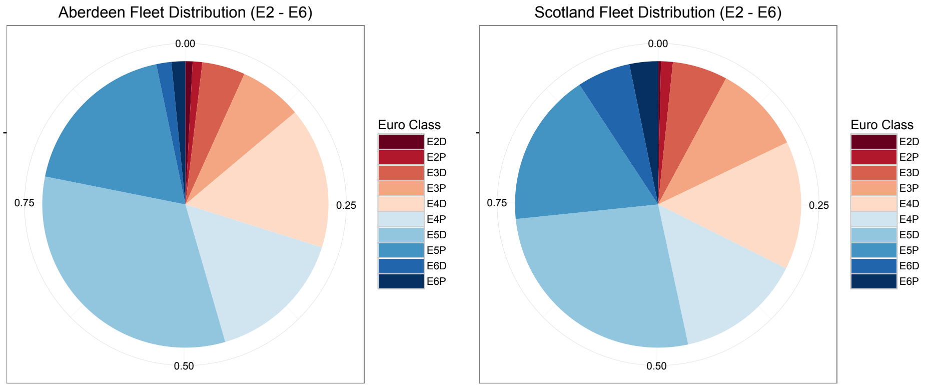

Figure 7.1: Pie chart showing the distribution of vehicle types in Aberdeen and London from Euro 2 - Euro 6.

Along with the base scenario and the future projections two additional fleet distributions are modelled to simulate the introduction of an ULEZ. The *Minimum Spend Scenario* (MSS) and the *Maximum Spend Scenario* (XSS) are both tested. The minimum spend scenario tests the scenario where people buy the cheapest vehicle they can afford that meets the criteria for the ULEZ, constrained in this case by vehicle age. A Euro 4 petrol vehicle is cheaper than a Euro 6 diesel vehicle. The maximum spend scenario asserts that people have a natural tendency towards a fuel type, either petrol or diesel, and that they buy a more modern Euro 6 vehicle and maintain their preference for fuel type. The rules for the two scenarios are detailed explicitly in the following two sets of bullet points:

- *Minimum Spend Scenario*
- All Euro 0 - 3 petrol vehicles are replaced by Euro 4 petrol
- All Euro 4 - 6 petrol vehicles remain the same
- All Euro 0 - 5 diesel vehicles are replaced by Euro 4 petrol
- All Euro 6 diesel remain the same

- *Maximum Spend Scenario*
- All Euro 0 - 3 petrol vehicles are replaced by Euro 6 petrol
- All Euro 4 - 6 petrol vehicles remain the same
- All Euro 0 - 5 diesel vehicles are replaced by Euro 6 diesel
- All Euro 6 diesel vehicles remain the same

These two purchasing scenarios represent two extremes that could be observed when legislation is introduced. The reality of the purchasing situation is likely to lie somewhere between these two cases. They do not take into account any decrease in vehicle number caused by changes in mode choice to either public transport, cycling or pedestrian. No difference route choice or time of day is modelled either. To attempt to mitigate this the model reports the average emission factor of a vehicle in the zone and these can be scaled up or down depending on the change in road network usage observed. For example if half the vehicle kilometres were travelled after the introduction of an ULEZ compared to before then the emission factor would stay the same per vehicle but the total emission of the fleet would be scaled by a factor of 0.5 alongside any additional gain by cleaner technology. These effects are beyond the scope of this thesis however the results obtained here would be useful to anyone attempting to ask this question in the future. The results are folded together to produce an aggregated emission factor per vehicle in the same way that the results in Section 6.4 are presented.

7.2.2 Estimating the Emission Factors

The base remote sensing data is that collected in the Aberdeen survey (Section 3.4.4). The ratio of $NO : CO_2$ is measured and the total NO_X emission factor for each vehicle is calculated using Equation 3.5. The CO_2 grams per kilometre were calculated using vehicle tracking data also conducted in Aberdeen at the same time as the remote sensing survey. The speed and acceleration profile was processed using PHEM to produce a CO_2 emission factor. The CO_2 emission factor was calculated for each vehicle fuel and euro class subset using the whole network approximation in Tate (2016). This method for estimating the CO_2 emission is used as it best represents the real driving environment in Aberdeen.

The model is run in the same way as is described in Section 6.4 with a number of adjustments. The fleets are changed to reflect Section 7.2.1 and the number of vehicles simulated is increased from 10000 to 24017, more representative of the number of vehicles measured in Aberdeen. This number was increased from Section 6.4 as fewer simulation runs were required and therefore was less computationally intensive. The baseline, MSS and XSS fleet distributions were each modelled and each fleet distribution model was repeated 10 times. As discussed in Section 5.4.2 the off model vehicles were considered and modelled using the Failed Catalyst (FC) approach. The aggregated emission value per passenger car for the 10 runs and the mean value is presented in Table 7.1 alongside the fractional change for the mean values of each scenario. The year required to achieve similar results (Control Year) is taken from the do-nothing cases in Section 6.4.

Run	Base [$gkm^{-}1$]	Maximum Spend [$gkm^{-}1$]	Minimum Spend [$gkm^{-}1$]
R1	0.729	0.468	0.309
R2	0.726	0.473	0.311
R3	0.733	0.469	0.310
R4	0.735	0.474	0.310
R5	0.733	0.471	0.311
R6	0.726	0.472	0.312
R7	0.718	0.472	0.312
R8	0.725	0.480	0.312
R9	0.720	0.477	0.311
R10	0.731	0.472	0.312
Mean	0.727	0.473	0.311
Base Change[%]	1.000	0.650	0.428
Control Year	-	2025 +	2025 +

Table 7.1: Aggregated emission factor [gkm^{-1}] per vehicle for the three ULEZ scenarios in Aberdeen

The results show that to achieve the worst case scenario, XSS, the natural fleet evolution in Scotland would have to wait until beyond 2025 before the the fleet could expect to produce the similar levels of decrease in aggregated emission factor if it were ever to be achieved reading the results from Table 6.7. The same is true of the minimum spend scenario however the additional 20% reduction is not close to any of the projections achieved in Section 6.4. This result is counter-intuitive as one would expect that the newer vehicles would be cleaner however the Euro 4 petrol cars, cheaper vehicles than the Euro 6 diesel cars, are the cleaner choice and result in less emissions. Getting people to convert from diesel to petrol may be a difficult task and the assumption in this scenario that they did is a big one however given the considerable difference between the two average emission factor reported for each scenario it seems like an attempt to turn people to these types of vehicles might be desirable. There is also an increase in variance between the individual runs in the XSS scenario. This is likely to be because of the greater number of off-model vehicles produced from the scenario, especially Euro 6 diesel vehicles. As is discussed in Section 6.4 the projected values are likely to be a best case scenario as vehicle ageing is not taken into account when deriving the off-model fleet fraction during calculation. Furthermore the Scottish vehicle predictions may still be lower than what might be expected in Aberdeen if the fleet is allowed to evolve naturally. The significantly higher number of Euro 5 diesels, a subset of the fleet that has high NO_X emissions compared to others, mean that locally the aggregated emission factor per vehicle may well be higher in Aberdeen than across Scotland as a whole. It is likely that it would take even longer than until 2025 to achieve the same results as could be achieved in a much shorter time by the introduction of an ULEZ.

7.2.3 Discussion

It has been shown that the introduction of an ULEZ in Aberdeen may significantly reduce the total NO_X emissions from passenger cars by between $\approx$ 35% and $\approx$ 57% depending on the choices drivers make when replacing their vehicles. These reductions are considerably greater than those which could be achieved by allowing the fleet to evolve over time naturally Given the current fleet projections it would take until at least 2025 for the effects of the ULEZ to be matched with reality. It is likely that given the tendency for Aberdeen car owners to buy an unusually high number of Euro 5 diesels compared to the rest of Scotland, possibly due to the higher fraction of company cars here than throughout the rest of the country, the time for the aggregated emission per vehicle to fall to that which would be expected after the introduction of the ULEZ will be even longer.

This analysis strengthens the case for adopting ULEZ's in urban environments to reduce NO_X. The results shows that over time it is unlikely that the fleet will adjust itself to reduce the total NO_X emission in urban environments. For policy makers that are serious about reducing the ambient air quality levels an intervention is the most effective solution despite the potential sources of conflict. The ULEZ restrictions that will be in place in London from 2020 should result in noticeable reductions in air pollution emitted by vehicles as long as the off model failed catalyst vehicle fraction remains stable. Given that in diesel cars this feature only occurs in Euro 6 vehicles then it is unlikely to be a feature of ageing and could be related to vehicle emissions systems intentionally being switched off. If this is the case the off-model fraction may remain stable into the future however future work must be done to assess the validity of this hypothesis. The off-model fleet picture in petrol vehicles is less clear as all classes are affected by it to some point. Further work should be done to better understand how the distribution of emissions from these vehicles change and if the problem appears to get worse then the projections should be adapted to take into account the new data.

The results presented in this section also show the limitations of Ultra Low emission zones with the best case scenarios only being comparable to a Euro 4 diesel vehicle. Furthermore no attempt has been made to model the contributions of LCVs (Section 7.3), HCVs and public transport. No additional attempts have been made to measure the impact of taxi vehicles (Section 7.4). These vehicles make up a significant fraction of the urban fleet and will have a non-trivial impact on the total emission factors. The failure to account for these vehicles is due to additional complexities related to LCVs that will be discussed in Section 7.3. In future work these vehicles must be considered if the projections are to represent anything other than a best case scenario however detailed information about the journey split is required in addition to the measurements of vehicle emissions. The introduction of new technology may still be required to further reduce the total NO_X emitted into urban environments. The total NO_X generated locally by electric vehicles will be zero and petrol hybrid powered vehicles ought to emit less than standard petrol vehicles. Testing these hypotheses and taking steps to further expedite the uptake of these vehicles into, and removing a higher number of traditionally powered vehicles from, the fleet ought to lead to further reductions in NO_X emission.

7.3 Impact of Light Commercial Vehicles in Urban Environments

The Aberdeen survey could have presented an opportunity to study Euro 6 light commercial vehicles and begin to make an assessment on their effectiveness in reducing air pollution. The dates for introduction of Euro 6 LCVs are described in Table 2.4.4 and some type NI vehicles might have been observed along with any other early adopters in the NII and NIII classes. After the survey was completed the total number of Euro 6 LCV's observed was zero and no emissions conclusions could be drawn. The large number of LCV's observed after the date of type approval for Euro 6 LCV NI class vehicles allowed an observation of how the changeover period between Euro 6 type approval and date of last registration would be handled by different manufacturers and also allowed an observation of the lifetime of the Euro 5 fleet to be made from first type approval to the introduction of new technology. Along with observing the characteristics of the fleet changeover it was possible to conduct a study of the emissions of these vehicles. Data was extrapolated over the whole network as well as a more simplistic snapshot that would normally be observed using an RSD by integrating vehicle tracking (Wyatt *et al.*, 2014) into the calculation of the CO_2 and hence NO_X emission factor. Data was collected using the standard on road setup and the sites are described in Section 3.4.4.

7.3.1 Introduction

Light Commercial Vehicles (LCVs) make up 9.1% of the registered vehicle fleet (Browne *et al.*, 2010; ICCT, 2015) and are responsible for even more journeys. They have a lifetime of approximately 5 - 8 years due to their intense usage. The non-trivial contribution these vehicles make to the total number of journeys made and their hours of operation means that it is important to understand the contribution made by LCVs to the total NOx in urban street environments. LCVs are subject to different mileage and increased loading compared to passenger cars however they are only occasionally tested in laboratories. LCV tests look at a small number of vehicles under controlled test procedures (Joumard *et al.*, 2003). Given their importance in the context of emissions in urban environments it is important to understand their contribution to the emissions inventory.

The type approval date for Euro 5 vehicles is September 2009 however newly registered LCVs do not have to meet Euro 5 standards until January 2011 for LCV-NI class and January 2012 for LCV-NII and LCV-NIII class vehicles. End of line legislation provides an additional mechanism for registering old vehicles. Either 30% of vehicles taken into use in Norway in the previous 12 months or 100, which ever is greater, can be registered in Norway. These vehicles may be finding their way into the UK fleet. It is not clear from the collected data how these vehicles are appearing in the fleet however they are there. The result of this legislation is that the new additions to the fleet after the type approval date may not necessarily change as quickly in response to the legislation would be desirable.

The RSD measurements along with the meta-data added through capturing of license plate data can allow monitoring of both the uptake of the new vehicles into the fleet and make an assessment of their total emissions. This methodology means that an accurate assessment of the uptake rate of vehicles and their emission can be used to better inform modellers and policy makers meaning that they can make more well informed decisions about policy implementation.

The Euro 6 legislation was introduced in September 2014 and an assessment of the impact is already possible for passenger cars using RSD measurements. At the time of writing it is still too early to assess the change in total fleet characteristics due to introduction of Euro 6 emission limit values however it is likely that policy regarding introduction of vehicles from various manufacturers will not differ too much from legislation to legislation. It is now possible to look at close to the whole lifetime of the Euro 5 legislation using remote sensing data collected in Aberdeen in 2015 and use this information to inform assumptions about the rate of uptake of new technology in the years following the introduction of the Euro 6 legislation.

The aims of this experiment were to attempt to describe the uptake rate of new LCVs into the fleet by looking back at the Euro 4 to Euro 5 fleet evolution and to assess how effective the Euro 5 legislation had been at reducing vehicle emissions and to compare them to the Euro 4 legislation.

7.3.2 Emission Factor Calculation

The same method used to calculate NO_X emission factors for passenger cars described in Section 3.3.2. A total NO_X emission factor is calculated for each vehicle by assigning it a fractional NO_2 value as measured in Carslaw & Rhys-Tyler (2013). Use of the remotely sensed fractional NO_2 values has been validated in Chapter 4 and can be considered the best available methodology for estimating f_{NO2} and is therefore used whenever possible. The total emission factor is calculated using Equation 3.5 where CO_2 is the urban cold manufacturer rating value of the vehicle. The CO_2 emission factor is calculated as shown in Section 3.3.2. The base CO_2 is taken as the cold urban rating, a rating that only covers urban driving, and uplifted by a scale factor of 1.23 to account for the differences between manufacturer rating and real world performance (Kadijk *et al.*, 2015).

To better understand the the LCV emission factor extrapolated over the entire Aberdeen network a Ford Fiesta hatchback was fitted with a VBox II Lite `(www.velocitybox.co.uk)` GPS and CAN interface (CAN002 module) instrument/ data logger sampling at 10 Hz. The position and road speed of the vehicle were recorded. The vehicle tracking survey took place on the 22nd, 24th and 25th of April 2015. The routes taken are described in the full report to Aberdeen City Council (Tate, 2016). The vehicle tracking is limited to one driver and it is assumed that this driver drives in a way consistent with the vehicles to be measured. The gradient is calculated by fitting the position track to a digital terrain model (DTM) http://landmap.mimas.ac.uk/. The purpose of using vehicle tracking is to provide a range of vehicle power demands that are more representative of the network as a whole and validate and expand on the results obtained by the RSD. The speed and gradient along with vehicle parameters are used as inputs to the passenger car and heavy vehicle emissions model (PHEM) (Hausberger, 2010). The vehicle mass, frontal area and engine power were sourced from the vehicle metadata collected by the RSD and the average vehicle payloads were specified as NI = 229 kg, NII = 382 kg, NIII = 478 kg (DfT, 2010). The CO_2 emission factor was calculated using PHEM based on these inputs and this CO_2 emission factor was substituted into Equation 3.5. The uplifted manufacturer rated CO_2 value was used to give an instantaneous emission factor measured at the remote sensing device. The manufacturer emission factor allows the greater variance between makes and models to be incorporated in the results but the PHEM derived CO_2 emission allows for extrapolation over the whole network. It is hypothesised that the two results will be similar.

7.3.3 Emission Factor Results

The total number of LCVs measured was 3511 and represented 14.2% of the total measurements. The total number of Euro 5 LCVs was 1895 with 96, 487 and 1312 being of class NI, NII or NIII respectively. Drive cycle tested Carbon Dioxide values were available for 90.64% of Euro 5 LCVs of all classes, or by class 92% for NI, 91% for NII and 86% for NIII. Measurements where no CO2 values were available were discarded. Of the remaining measurements, 97.8% of NI, 97.2% of NII and 96.1% of NIII measurements for the NOx emission factor in grams per kilometre were found to be over the limit values required for the Euro 5 type approval. Repeating the analysis using the average PHEM CO2 emission factors extrapolated over the whole network results in 96.9% of NI, 96.7% of NII and 93.4% of NIII LCVs being over the limit values. These limit values are 0.180, 0.235 and

0.280 grams per kilometre respectively. The two CO_2 measurements produce similar results and suggest that using the PHEM CO_2 values alone does not lose too much aggregate the data in such a way that significant real changes could not be observed.

The RSD and metadata collection methodology has allowed analysis by manufacturer to be performed across the fleet of LCVs. The distribution of the measurements, shown in Figure 7.2, varies significantly between manufacturers. Some manufacturers are able to produce many vehicles that whilst over the limit are still relatively closely distributed around it, for example Ford, whereas other manufacturers, such as Vauxhall, distribution of emissions are spread out to both extremes of the scale. The distributions of NO_X emission factors subset by manufacturer are presented in Figure 7.2 for NI, NII and NIII consecutively. Grey histogram bars show the distribution of emission factors as calculated using the uplifted manufacturer rated CO2 values and red, blue and green histogram bars show the emission factors calculated using the PHEM CO_2 emission factors for NI, NII and NIII respectively. The legislated limit value for Euro 5 vehicles is indicated as a dashed red line in each separate histogram. It is beyond the scope of the RSD survey to understand what the different solutions and applications of technology are or to suggest ways that manufacturers could improve their real world emissions, only that a difference is clearly visible in the data. This further complicates any attempt to remove high emitters as manufacturers who are able to effectively reduce their NOX emission may be unfairly penalised or those that are not performing as well may be allowed to get away with emitting more NOX than others.

These results show that the emissions produced by these vehicles as of 2015 are emitting more NO_X than their limit values allow to the environment. The results of this analysis also show that whilst no

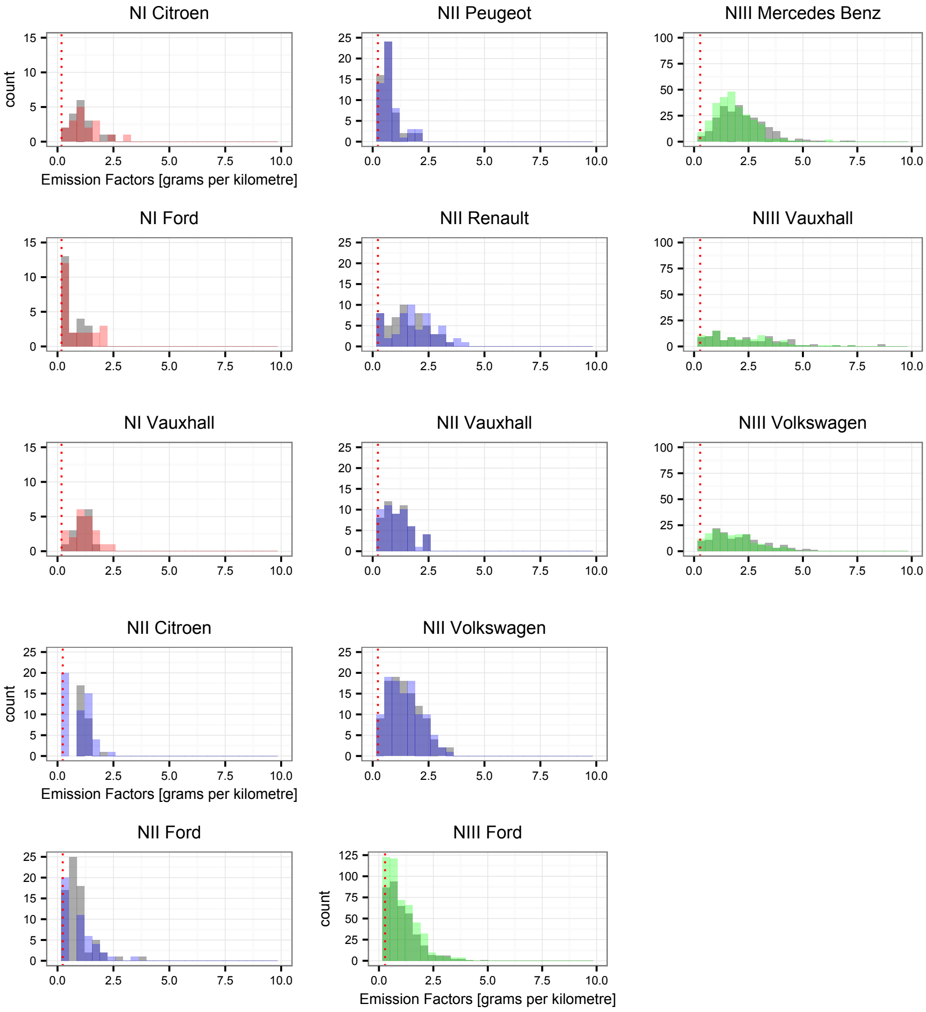

Figure 7.2: Distribution of NOx emission factors over a range of LCVs in Aberdeen.

manufacturer can claim to be under the limit values some manufacturers are producing vehicles that emit less NOx than others in the NII class and especially in the NIII class. In the NIII class for example Ford vehicles peak only slightly higher than the limit value whereas Mercedes, Vauxhall and Volkswagen as well as potentially Renault and Vauxhall, which have a smaller sample size, have both peaks and spreads that are more in excess of the limit value and covering much more of the range of emission factors than the better manufacturers. This level of complexity means that modelling the behaviour of this subsection of the fleet is more complex than modelling the passenger car subsection. A two-sample K-S test was applied to the four NIII manufacturers to test if they were likely to be from the same distribution. The only two sets with $p > 0.05$ were Vauxhall and Volkswagen ($p = 0.051$). The next two closest subsets were Mercedes-Benz and both Vauxhall and Volkswagen ($p = 0.016$). The distribution of emissions in the Ford fleet is significantly different from all the rest of the manufacturer subsets ($p \approx 10^{-15}$). This suggests that there is a significant difference between manufacturers and therefore that they cannot be convolved into one distribution for modelling purposes in the same way that the passenger cars have been

7.3.4 Introduction of New LCVs Into The Fleet

Analysis of the date of first registration for each vehicle was performed to better understand how LCVs were introduced to the vehicle fleet both in the run up to and after the introduction of new legislation. Given the 5-8 year lifetime of a fleet vehicle it is assumed that the majority of vehicles registered since 2007 and almost all registered since 2010 were still present on the road when the observations were made. Given that September 2010 was the introduction date for new type approval the time period of interest should be well represented by the vehicles measured on the road. The measurements were taken after

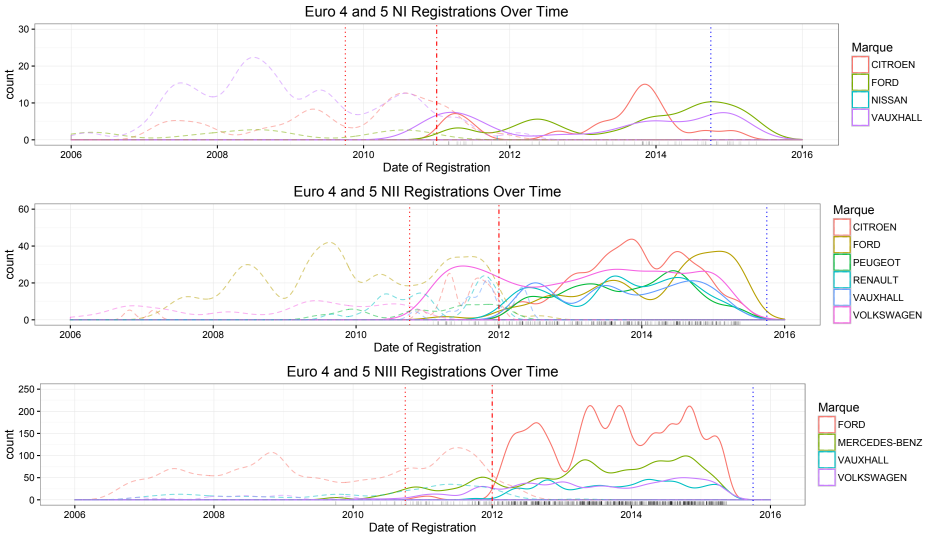

Figure 7.3: Uptake of new LCV's into the Aberdeen market for most popular marques.

the introduction of Euro 6 (See Table 2.4.4) vehicles but no vehicles measured were approved to the standard of the new legislation.

The introduction timelines for LCVs fitted with Euro 4 (dashed) and Euro 5 (solid) technology are shown in Figure 7.3. The technology is presented in frequency polygon form with an adjustment resolution of 0.4 years (0.6 for NII Euro 5 for reasons of clarity). Additionally a rug is presented to show individual vehicles day of registration. Darker grey indicates more vehicles registered on that day. Dashed red lines indicate the last type approval date and the last registration date from left to right respectively. The blue dashed line indicates the last type approval date for Euro 5. The rate of introduction of new technology is not as clear-cut as might be expected given the nature of the legislation. There are some manufacturers who get their new stock on the road considerably quicker than others. Without detailed information about the logistics systems that each manufacturer is using to fulfil its orders the reasons for this cannot be known. The difference between manufacturers introduction of new vehicles means that care must be taken when attempting to restrict vehicles. Simply specifying a year of registration is not enough to ensure that all vehicles meet the required emissions standards and explicit statements about the Euro class of a vehicle must be used. Based on this analysis the contribution of LCVs to the roadside concentration of pollution should not expect to decrease alongside new type approval legislation. A lag period of one year between the introduction of the Euro 5 type approval and more than just a small number of newly registered vehicles meeting the Euro 5 emissions limits has been observed. During this time little to no decrease in roadside concentration would be expected due to new LCVs.

The analysis undertaken previously is expanded to understand the strategies that different manufacturers use during the period of transition between Euro class legislations. Given the freedom that is afforded these manufacturers by the legislation some variance would be expected. Figure 7.3 shows the uptake rate separated by manufacturer for the period 2006 to 2016. The two dashed lines indicate the transition period with the first one representing the type approval date and the second one representing the new registrations date with manufacturers indicated by different colours.

7.3.5 Conclusions

The implications of the results of this research are important from a policy makers point of view. Firstly it has been shown that Euro 5 vehicles are emitting NO_X at a level considerably higher than the legislated limit values. The limit values cannot be used for any sort of policy-making decisions moving forward as they produce a significant underestimate. The range of observed values across LCV class types (NI - NIII) is mostly consistent therefore banning specific types of LCV would not significantly change the total emitted NO_X. The inter-marque effectiveness at reducing NO_X is also striking with some significant differences observed between different manufacturers predominantly in the NIII class. The implications of this result are that there are some system design solutions that are more effective than others at removing the NO_X but that not all manufacturers have been as successful at optimising their technological strategy. This result provides a complication for policy makers as it means that some manufacturers vehicles may be unfairly penalised or other manufacturers may be able to avoid optimising their technology if simple legislative interventions to reduce the impact of LCVs are implemented.

The impact of Euro 6 LCVs has not been assessed, as despite the measurements being taken approximately 8 months after the new type approval date there were no examples of Euro 6 LCVs present in the Aberdeen fleet measured by the RSD. The lack of vehicles meeting the new limit values further complicates the picture in imposing restrictions on vehicles to limit the total NO_X emissions as limitations based on the type approval year cannot be trusted to remove all old technology. This result is surprising as Euro 6 cars have been observed in reasonably high numbers since and even prior to the introduction of the new limit values.

It has also been shown that the last change of legislation from Euro 4 to Euro 5 did not produce immediate change in technology that was introduced into the fleet. Whilst it remains too early to confirm it appears that this lag between legislation and technology introduction is happening with Euro 6 technology as well. Whilst it would be unrealistic to say that the fleet of Aberdeen can be considered representative of the whole UK fleet without further investigation nor that the sites monitored measured every LCV in Aberdeen the initial stages of the Euro 6 legislation period are similar to the initial stages of the Euro 5 legislation. Any attempt to reduce high NOX emitting LCVs from a low emission zone should account for this lag.

The work done in Section 6.4 to project the future fleet emissions needs to be extended to include LCVs as they make up a non-trivial fraction of the fleet and are emitting more than their type approval values of NO_X. The variation between manufacturers emissions is greater than observed between different manufacturers of passenger cars (Section 7.5) and this makes it more difficult to integrate LCVs into the model. It remains to be seen whether or not the behaviour of the whole fleet can be convolved into one distribution. Ford NIII vehicles behave very differently from Vauxhall NIII vehicles both on the road and in statistical descriptions and it remains to be seen whether or not the LCV fleet can be modelled in the same way as the passenger car fleet can be or whether additional steps are required. Given the success of the model proposed in Chapter 6 and the number of LCV's in the fleet it is worth further investigating and attempting to integrate the LCV fleet into the passenger car fleet model.

The rate of introduction of new technology into the vehicles on the forecourt has been observed using license plate information and has shown to vary quite considerably depending on the manufacturer with some being able to get their newest products into the fleet very rapidly whilst other manufacturers are waiting until the last possible moment to introduce theirs. This presents a problem for policy makers because it means that not all LCVs registered in (for example) 2012 are working to equivalent standards. Some may be Euro 4 whilst others may be Euro 5. Any intervention to limit the access of old vehicles or promote access for new vehicles in a low emission zone scenario therefore risks either unfairly punishing a manufacturer who has introduced new technology early or encourages manufacturers whos vehicles are running to an older specification of legislation to not introduce their new technology as soon as possible. In practice the difference between Euro 4 and Euro 5 is limited so the ambiguity caused by different introduction times is largely inconsequential however if Euro 6 technology can be shown to significantly reduce NOX in comparison to

Euro 5 it is important to understand how to incentivise the uptake of the new technology not just new vehicles that only meet the old standards.

In continuation of this study a measurement of the emissions performance of Euro 6 LCVs is critical as no assessment is currently possible. At the time of writing the type approval deadline has passed for Euro 5 M class vehicles and NI class vehicles (see Table 2.4.4) however the vehicle registration deadline has not yet passed for Euro 5 NII and NIII class vehicle. The vehicle registration deadline for Euro 5 NII and NIII vehicles is September 2016. As time progresses, the flexibilities and other derogations run out and the fleet turnover continues to evolve naturally the likelihood of measuring these vehicles using the RSD increases. The introduction rate of Euro 6 vehicles should also be monitored. As these deadlines pass the impact of the Euro 6 legislation will be seen in these vehicles and their rate of uptake can be observed in real time by repeated RSD studies from May 2016 and further into the future. It is highly recommended that this study is undertaken in the future.

7.4 Differences Between Taxis and Private Passenger Cars

7.4.1 Introduction

Taxis represent a significant number of vehicles in the fleet, especially in urban environments. The total number of taxis registered in Leeds is 4387, the majority (87.7% or 3851) being typical passenger cars such as the Skoda Octavia (947 registered), Toyota Avensis (511 registered) and Vauxhall Vectra (399 registered). These vehicles are privately run and may not be hailed on the street. The remaining 12% are 'Hackney Carriages'. These vehicles are typically older, heavier, diesel powered vehicles specifically designed to be taxis such as the Black Cabs in London, with higher passenger capacity than PHVs. Taxis represent a significant proportion of the total journeys completed especially during the night where they can account for 35 percent of the journeys, decreasing to $5-10\%$ during the peak hours (LCC, 2015; Riley, 2016; Rogerson, 2013).

The typical operation cycle of a PHV taxi is different from a equivalent privately owned vehicle. PHV taxis may often run back to back 12 hour shifts and cover 10×10^5 kilometres a year compared to the UK average of 1.3×10^4 kilometres (DfT, 2015). Furthermore the PHV use cycle may not include the high-speed periods required to trigger the rich running engine mode necessary to regenerate the catalyst (EA, 2013) and to maintain it at operating temperature if traffic becomes congested (Herreros *et al.*, 2014). If taxis are operating significantly differently to private hire vehicles and not in their designed optimised windows it is plausible that the vehicles may exhibit different emissions characteristics from other vehicles that are being run closer to the optimised windows. Given the large contribution these vehicles make to the total fleet mileage it is important to understand how to model them in relation to the rest of the fleet in line with the research goals outlined in Section 1.3 so that fleet interventions that include a shift from privately owned vehicles to taxis is well understood.

Current modelling techniques do not account for the differences between a private hire vehicle, a privately owned vehicle or a Hackney carriage. Taxis are typically not considered discretely in all but the London Atmospheric Emissions Inventory (LAEI), just part of the passenger fleet. If there is a significant difference between these two sections of the fleet then failure to distinguish between the two in terms of their emissions may lead to a misestimate of the pollution inventory derived from models. Kerbside remote sensing of vehicles offers a rare chance to observe a range of vehicles in-situ, and in this case directly measure members of the taxi fleet, which have been exposed to a range of operational histories hence improving the clarity of understanding. Substituting environment, drivers and vehicle characteristics control for volume and breadth of data means that an opportunity to compare different elements of the fleet in a new way is possible.

The NO emissions of the fleets of taxis in both Sheffield and Cambridge were measured in-situ . Taxis in the fleet were identified using the license plate database provided by the respective city councils. This is the first time taxis have been directly measured on the road using a remote sensing device along with the meta-data required to compare them to other subsections of the vehicle fleet. Their impact on the urban environment and their pollution compared to other subsections of the fleet is investigated both quantitatively and qualitatively in this section and two methods for reducing the total NO_X emissions of taxis are discussed. The results of this research are of interest to emissions modellers who may wish to up-lift the impact of taxis compared to regular vehicles, and help policy makers develop a clearer understanding of the sources of air pollution.

7.4.2 Methodology

In this experiment the data sets from Sheffield (Section 3.4.1) and Cambridge (Section 3.4.2) were combined. This research was performed before the Aberdeen study was conducted and represented the best range of data available at the time. The taxi registration database was made available by both Sheffield and Cambridge city councils. The vehicles were subsetted into one of three categories. Privately run vehicles (PCs), private hire vehicles (PHVs) which are typical fleet vehicles but registered to private hire taxi companies and Hackney Carriages (HCK) which are bespoke designed taxi vehicles.

The privately owned car data was divided up into sub-sets based on Euro class and fuel type that corresponded to the PHV and HCK fleets. Euro 3 and 4 Diesel vehicles were compared to Hackney Carriage vehicles and Euro 4 Diesel vehicles were compared to the PHV fleet. Taking sub-sets of the fleet eliminate both the potentially highly emitting lower and higher Euro 2 and 5 vehicles and ensure that as best as possible a like for like comparison has been performed. Of chief interest in this experiment are vehicles that are either Private Hire or pre-booked taxi Vehicles (PHVs), Hackney Carriage or hailed taxis and Privately owned vehicles that are representative of these subgroups.

Fractional NO_2 is calculated using the method described in Section 4.3. No distinction is made between the taxis and privately owned vehicles when assigning the fractional NO_2. The CO_2 emission factor is calculated using PHEM for both instantaneous emissions and network wide emissions. The instantaneous emission factor is calculated using a new method. The speed and acceleration measured by the RSD was extrapolated to create two speed and acceleration points and these points are used as inputs to PHEM to calculate the total CO_2 emission. This methodology has proven to be particularly computationally difficult as PHEM is not designed to input and output files with this frequency and for this reason this experiment is the only instance in this thesis where it is used. Network wide CO_2 emission factors are derived using the same PHEM model but using vehicle tracking conducted over the Sheffield and Cambridge networks respectively, in line with work done in Section 7.3 and throughout the rest of this thesis.

7.4.3 Results

The RSD equipment was run for 20 days over 10 sites in Sheffield and Cambridge. Over the course of this over $50k$ vehicles were measured, $30k$ of these were passenger cars. A total of 13255 cars were selected for analysis from these data sets. Of these, 4.6% were Hackney

Fleet Section	Frequency
Private Diesel Car	11678
PHV Representative	4817
HCK Representative	7241
Private Hire Taxi	965
Hackney Carriage	612
Total	13255

Table 7.2: Number of taxis measured in Sheffield and Cambridge

Fleet Section	Mean NO $[gkm^{-1}]$	Mean NO_X $[gkm^{-1}]$
Private Diesel Car	0.19	0.26
PHV Representative	0.16	0.24
HCK Representative	0.18	0.25
Private Hire Taxi	0.23	0.33
Hackney Carriage	0.23	0.32

Table 7.3: Total NO and NO_X emitted by taxis in Sheffield and Cambridge

Carriage and 7% were private hire vehicles. The number of vehicles measured in each sub-set are shown in Table 7.2.

The total NO_X and the NO was calculated for each fleet subsection described in Table 7.2 are shown in Table 7.3. The results show that for both cases the taxi fleet is emitting more NO and NO_X than a privately owned fleet that is representative of the type of vehicle found in the taxi fleet by factors of 1.38 and 1.28 for private hire taxis and hackney carriages respectively and compared to the total fleet of diesel powered vehicles by a factor of 1.23.

The distribution of the total NO_X emission factors for PHVs and Hackney Carriage vehicles is presented in Figure 7.4 and Figure 7.5

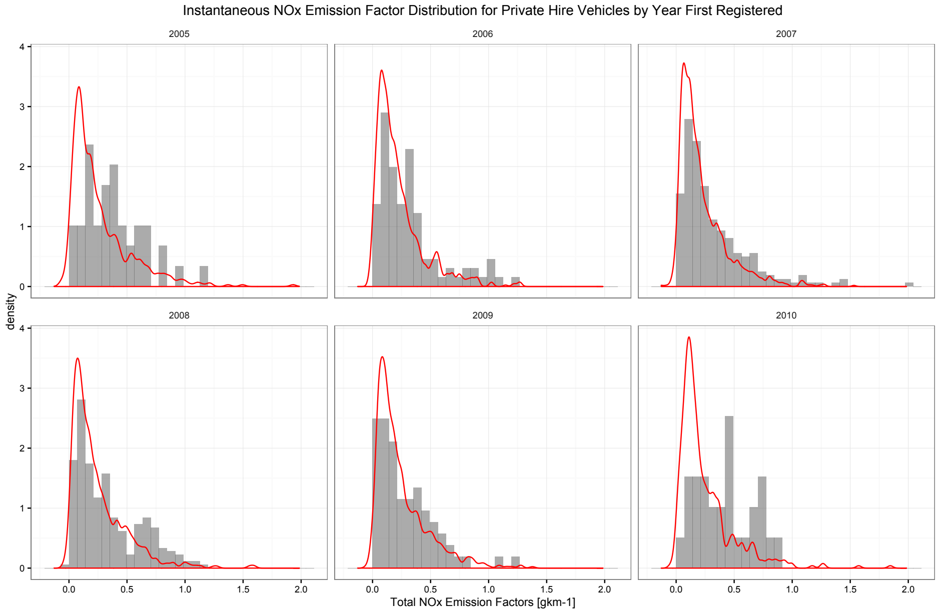

Figure 7.4: Distribution of PHV emission factors compared to the representative fleet

as a grey histogram. The red frequency polygon indicates the distribution of the representative fleet. This result shows that there is a difference between both taxi fleets and their representative privately owned vehicles. Due to age restrictions placed on the fleet the number of PHV taxis in 2011 and 2012 is not large enough to provide meaningful analysis.

A crude analysis of the PHV distributions in Figure 7.4 show that the reason for the increase in average emission values is due to a fatter tail caused by more extreme values however the distribution of emissions remains largely similar to the distribution of the representative privately owned fleet. The subset from 2010 show a different distribution to the private fleet however the sample size for this year is lower than for the other years so it is possible that if the sample size were to be larger the distributions of the two fleets would converge on a similar shape. The same analysis of the Hackney Carriage fleet shows

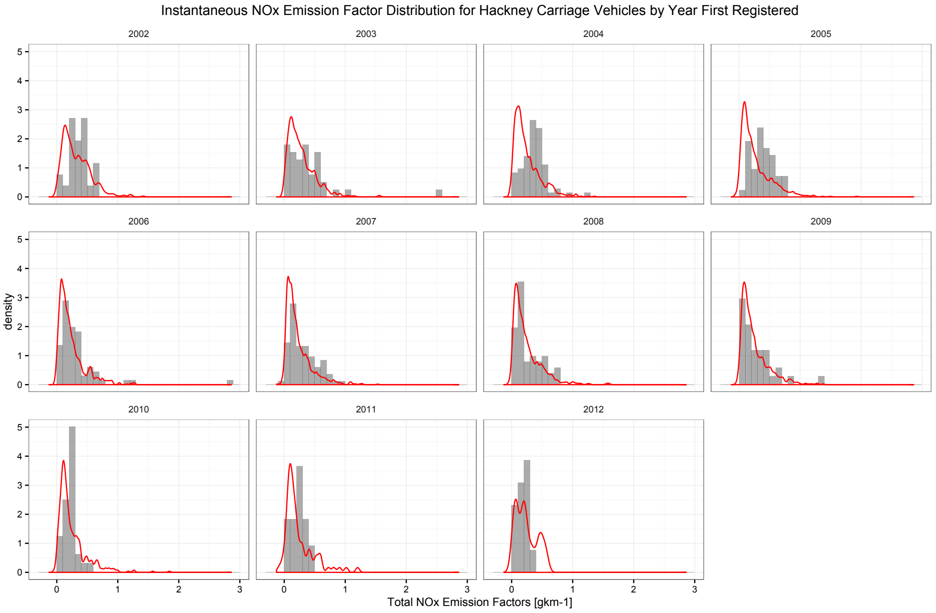

Figure 7.5: Distribution of Hackney Carriage emission factors compared to the representative fleet

a broadly similar picture although the number of extreme values observed in the earlier registered vehicles is notably higher than in the PHV fleet.

The distribution of all of the fleet subsets studied in this section are asymmetrical and follow a distribution more in line with an extreme value distribution (Chapter 5). Analysis of the NO_X emission factor distribution within the framework of an extreme value distribution can give a more rigorous description of the differences between the two fleets. The methodology for using the extreme value distribution applied to emission factors is described in Chapter 5. Five candidate distributions were selected for consideration and a Gumbel distribution (Gumbel, 1935) is selected to model the four data sets as it gives the best fit when examined using Q-Q plots. These plots are shown in Figure 7.6 and Figure7.7 for the taxi fleet and the fleet representative of taxis respectively. For valid comparison all the plots are translated

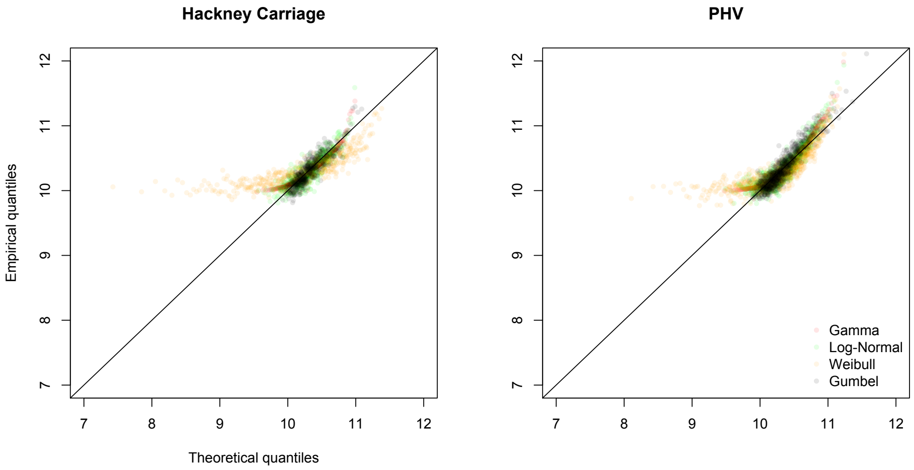

Figure 7.6: Q-Q plots of taxi fleet NO_X emission factors

$+10gkm^{-1}$ as some of the distributions require all positive numbers prior to fitting. The P-P plots of all the fleets are shown in Figure 7.8. Whilst the distribution is not a perfect representation of the fleet it is the best available solution for mathematically describing and hence quantitatively comparing different fleets.

The location parameter (a) and the scale parameter (b) correspond to the central point of the peak and the spread respectively. The fitted parameters and their standard error indicated by ± for each fleet subset are presented in Table 7.4. In the context of total fleet emissions the lower both of these parameters is the better.

These results show that both the peak in the distribution and the frequency of higher than modal emitting vehicles is greater in both the private hire section and the hackney carriage section of the fleet and significantly higher than when the whole diesel fleet is modelled. As described in Chapter 5 the location parameter is equivalent to the mode and the scale parameter is related to the number of vehicles in the tail. The fitted parameters from the Gumbel distribution (as described in Chapter 5 and used thereafter) are different from the mean

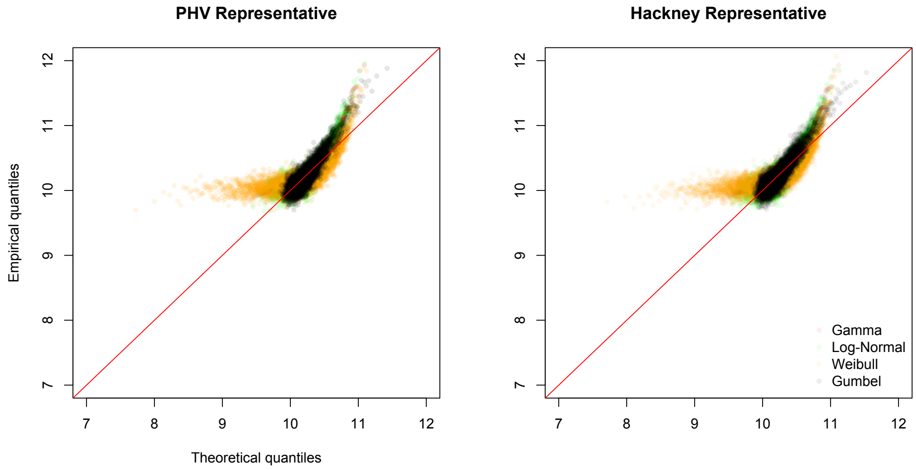

Figure 7.7: Q-Q plots of taxi fleet NO_X emission factors

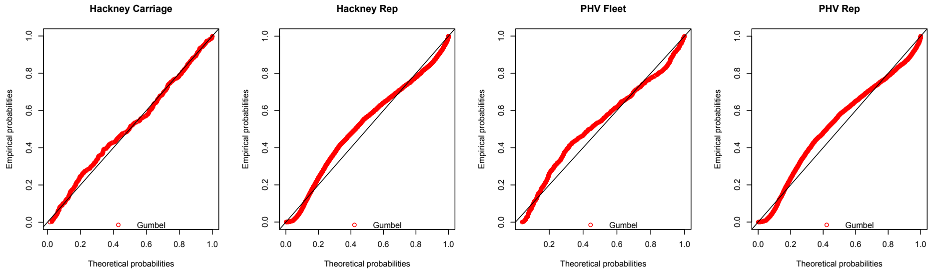

Figure 7.8: P-P plots of all vehicles' NO_X emission factors

Fleet Section	Location (a)	Scale (b)
PHV Representative	0.148 ± 0.002	0.139 ± 0.002
HCK Representative	0.156 ± 0.001	0.144 ± 0.001
PHV Taxi	0.212 ± 0.007	0.187 ± 0.006
HCK Taxi	0.218 ± 0.008	0.166 ± 0.006
All Diesel Cars	0.167 ± 0.001	0.149 ± 0.001
Euro 5 Diesel	0.180 ± 0.003	0.155 ± 0.002
All Petrol Cars	0.0401 ± 0.0005	0.0722 ± 0.0005

Table 7.4: Location and scale parameters for Gumbel distribution fits including standard error

and standard deviation suggesting that the parameters produced by fitting the clearly asymmetrical data to an asymmetrical distribution are better for analytically describing the fleet. This observation reinforces the conclusion that the Gumbel distribution is the best distribution for describing vehicle emissions drawn in 5. The poor ability of the Gamma distribution, most clearly visible in the PHV only Q-Q plot to describe this data is additional evidence to back up this conclusion.

7.4.4 Conclusions

It has been shown that taxis emit more total NO_X than their fleet equivalent vehicles that are privately owned. The remote sensing device methodology does not allow inspection of what the engine control unit is doing but a noticeable difference can be observed between the taxi fleet and the representative fleet. The difference between the fleet subsets has been shown in a quantitative way for the first time using remote sensing and it can be seen that taxi fleet emits both a higher modal amount of NO_X and that the frequency of higher emitting vehicles within the population is also greater than for the taxi fleet as a whole and have confirmed the initial hypothesis. Modernising the taxi fleet to Euro 5 diesel standard would not have a positive effect as they emit more total NO_X than the Euro 3 and Euro 4 private vehicles representative of the current taxi fleet. It is hypothesised that this is because the stricter emissions tests for Euro 5 than 3 or 4 means that the operation cycle for taxis is even further out of their optimal operating window and the result is higher NO_X emission. Incentive strategies to pull taxi operators towards petrol and petrol hybrid power would be more beneficial to NO_X reduction than pushing taxi operators towards more modern diesel powered vehicles. It is concluded that replacing both PHV taxis and hackney carriages with petrol or petrol hybrid as discussed further in Riley (2016) represent an opportunity to significantly reduce the NO_X emitted by taxis in city centres and other urban environments where they contribute a large proportion of the vehicle kilometres travelled.

7.5 Detecting Defeat Devices Using a Remote Sensing Device

7.5.1 Background Information

On 9 September 2015 the Environmental Protection Agency (EPA) gave a Notice of Violation of the Clean Air Act (CAA) to Volkswagen Group (VWG) stating that they had been using a device to circumvent the emissions tests, also known as a defeat device, on their 4 cylinder engine designated EA189 from 2009 to 2015. Approximately *five hundred thousand* vehicles in the USA were affected. The defeat device was able to sense when the vehicle was being tested and switch to an engine mode that produced less emissions and switch out of it on the road where better performance was required.

It has been the goal of VWG to supersede Toyota Motor Corporation (TMC) as the largest manufacturer of vehicles in the world. As of August 2014 VWG had a global market share of 11.1% compared to 11.6% market share possessed by TMC. VWG's solution to this problem was to break into the vehicle market in the USA. To this end a high performance low emission diesel vehicle was designed. It was believed that this would help to increase the market share VWG in the USA and propel the group towards it's goal of being the world leader in vehicle sales.

Whilst the EA189 engine passed all the type approval tests the lab results fell considerably short of the real world emissions and that a defeat device had been installed in the engine control unit. It later emerged that the defeat device had been installed across the whole spectrum of vehicles fitted with the EA189 engine worldwide, not just limited to the United states, bringing the total number of vehicles affected by the scandal to approximately eleven million (Times, 2015).

The implications of this scandal have been huge. Financially the stock value of VWG has dropped by approximately 40%, senior board members have resigned and the cost of fines and as of 15 March 2016 a 3.3 billion lawsuit has been filed against VWG by investors for failing to inform them quickly enough of events as they unfolded (`http://www.theverge.com/2016/3/15/11232062/volkswagen-diesel-emissions-lawsuit-germ` accessed 15 March 2016). Further lawsuits relating to the decrease in resale value amongst others have also been posited.

Environmentally vehicles are still emitting considerably more NO_X than the type approval limit values and the knock-on health impacts are being felt over a wide area with little sign of anything changing. The scandal has highlighted both the inadequacies of the testing procedure for diesel vehicles and the lengths that some manufacturers are willing to go to achieve a competitive advantage on both pricing and marketability of their products.

7.5.2 Remote Sensing Application

Previous to 2009 VWG diesel vehicles used a Turbo Direct Injection (TDI) system but after 2009 a computer controlled common rail Diesel Direct Injection Engine (DDIE) system using high pressure pumps manufactured by Bosch was used in the new EA189 engine. The benefits of this new injection configuration were better control of the fuel flow and better atomisation of the fuel leading to better fuel consumption and air to fuel ratio control leading to better control over emissions. The ECU system for the EA189 engine was able to access modes in the fuel pump that allowed very low emission when the vehicle was determined to be undergoing a test but reverting to less favourable emission levels when no test was detected. This allowed the vehicle to be marketed as a performance vehicle but still meeting the Tier 2 NO_X emissions limits. Use of these so called defeat device is illegal under the CAA.

In Europe the NEDC is considerably more favourable to manufacturers than the FTP75 drive cycle (Section 2.4) so if it was possible to detect the latter being conducted it would not be difficult to test for the former. With VWG admitting that the total scope was 11 million vehicles worldwide it would appear that the use of defeat devices was not limited to the USA but represents an attempt to defraud the global automotive market with no regard for the significant health issues resulting from NO_X emissions in urban environments.

With the precise nature of the testing procedure known well in advance it is possible that a sufficiently motivated line manager or engineer would be able to use the myriad of sensors on modern vehicles to be able to determine if the vehicle was being tested. An attempt was made by an unpaid enthusiast to reverse engineer the ECU for an EA189 diesel engine to see if a defeat device could be found. It was found that the ECU was tuned through distance time calculations as well as system temperature sensors to be able to know that it was on the NEDC test cycle and switch to an engine mode that would allow it to pass. The full technical description is beyond the scope of this thesis however the keynote speech demonstrating this is available online (`https://media.ccc.de/v/32c3-7331-the_exhaust_emissions_scandal_dieselgate`). If the admission of guilt was not convincing enough the routines that were uncovered by these enthusiasts is the smoking gun that proves not only that VWG was illegally passing the test but that it was exploiting the formulaic design of the test procedure to do so.

Remote sensing it's self cannot directly identify whether or not a defeat device is in use or has been used previously to pass a test however as has previously been mentioned throughout this thesis it can provide insight into the real driving performance of vehicles that have passed the NEDC. As the meta-data collected alongside vehicle emissions contains information about make and model it has been possible to not only check the allegations that VWG vehicles are polluting above and beyond the limit values set out in the Euro 5 legislation but also compare their emissions to those of similar vehicles produced by other manufacturers.

A number of research questions that were not of concern at the start of the thesis become topical when the VW scandal broke. The questions could however be investigated using the RSD data and vehicle metadata. This section seeks to answer the following questions questions: Do VWG vehicles fitted with the defeat device significantly breach the limit values specified by the Euro class regulations? Are VWG cars the only ones who are over the limit or is the problem systemic within the industry? What is the impact of these vehicles on the road?

7.5.3 Performance of VWG Vehicles

To ascertain whether or not VWG vehicles that use the EA189 engine emit more than the Euro 5 limit values in real driving environments all those vehicles fitted with that engine and isolatable in the vehicle database were selected. The selection criteria was as follows: All vehicles in the Aberdeen data set that were made by a VWG marque, Audi, Seat, Skoda or Volkswagen, were extracted from the data set as a whole. Those fitted with 2.0L EA189 engines were further extracted. Data for the 1.6L EA189 engine was also available however the sample size was small compared to the 2.0L dataset and not used. An emission factor was calculated using Equation 3.5 with the

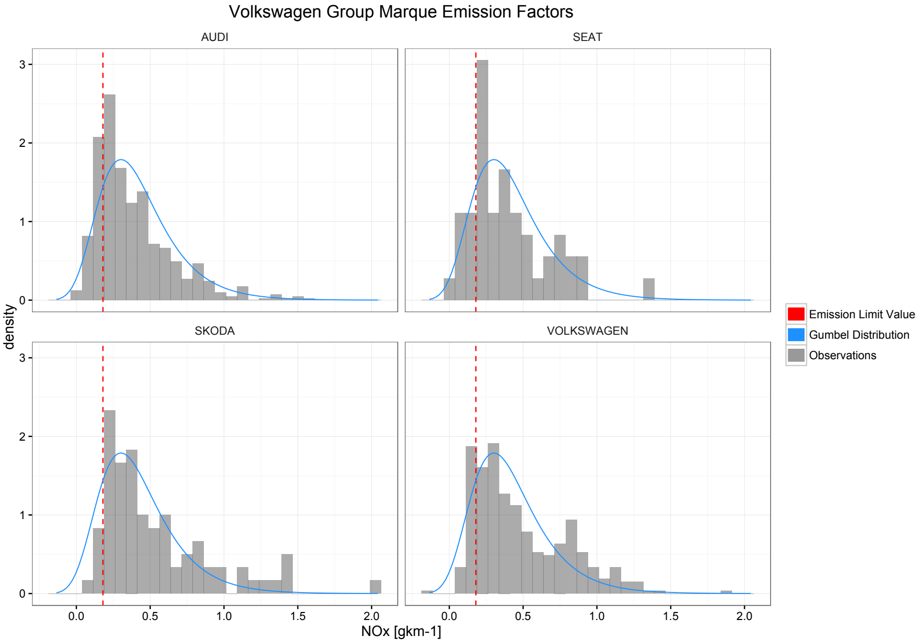

Figure 7.9: NO_X emission factors for all VWG marques

$f_{NO2} = 0.24$ value taken from Carslaw & Rhys-Tyler (2013) and the $CO_2 = 240gkm^{-1}$ value estimated using PHEM and vehicle mapping covering the whole of Aberdeen's road network (Tate, 2016). Using this method allows for a better idea of the vehicles' impact over a city rather than instantaneously. Choosing one CO_2 factor is more representative as only one type of vehicle is tested. The emission factors were binned and presented as a histogram in Figure 7.9 with a red dashed line representing the Euro 5 diesel drive cycle homologation limit of $0.180gkm^{-1}$. The data are plotted in a density histogram to allow comparison between the four different manufacturers despite there being significantly more Audi and Volkswagen marque vehicles than Skoda or Seat.

It is clear that the VWG vehicles are exceeding the limit values required for type approval. A Gumbel distribution is fitted to each marque individually and to the VWG data set as a whole. The results show that the VWG fleet as a whole has parameters of location (a) and scale (b) of $a = 0.386 \pm 0.009$ and $b = 0.263 \pm 0.007$. In comparison to this the results for Audi, Seat, Skoda and Volkswagen are $a = 0.35 \pm 0.01$ and $b = 0.229 \pm 0.008$, 0.36 ± 0.04 and 0.25 ± 0.03, 0.48 ± 0.04 and 0.31 ± 0.03, and 0.44 ± 0.02 and 0.30 ± 0.01 respectively. There is some natural variation between the fitted distribution parameters between models but the errors associated with these parameters, calculated as the standard error by the fitting algorithm (Delignette-Muller & Dutang, 2015), suggests that across the four marques these values are consistent with one another. This confirms expectations as all the vehicles are all running on the same engine and similar chassis platform.

The difference between different models of the same marque are also of interest. The Volkswagen and Audi marques are subset down to individual models because there are a large number of observations available. Skoda and Seat are not analysed in this way as their sample size is too small when models are considered individually. The following models were removed from the Volkswagen set as the sample size was too small ($n < 10$): Beetle, Golf Plus, Jetta, Touran and Transporter (car derived). The emission factors are presented in Figure 7.10 and Figure 7.11 in the same style as Figure 7.9.

A number of different Audi models were observed in the fleet. There is a degree of consistency in emission factors across the models with some natural variation observed. The A1 ($n = 15$) and TT ($n = 10$) sample sizes are small compared to the rest of the Audi models and this may account for the high standard errors that are observed on their distribution functions presented in Table 7.5. Overall the individual models for Audi are consistent with one another.

As with the Audi model analysis the Volkswagen analysis is consistent across models. The exception to this is the Tiguan model that exhibits higher than expected peak emissions as shown in Table 7.5.

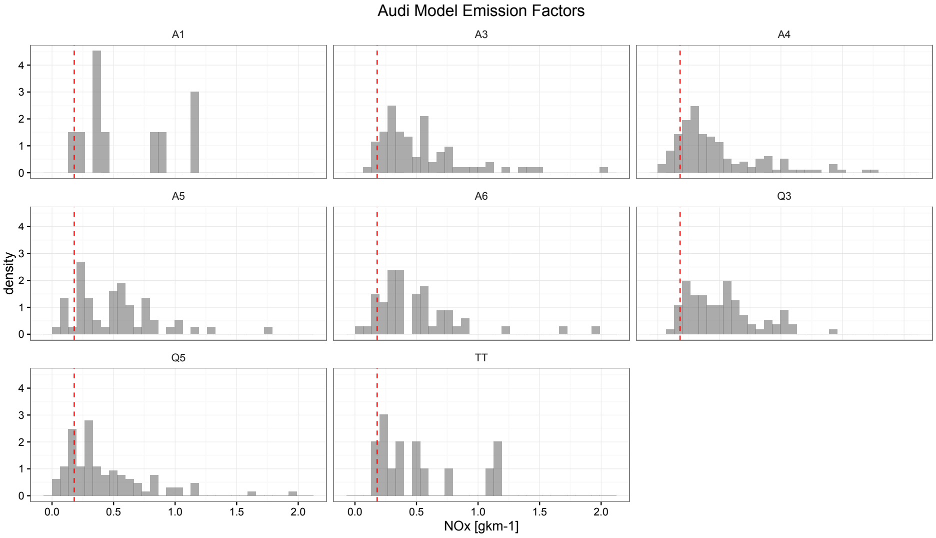

Figure 7.10: NO_X emission factors for Audi models

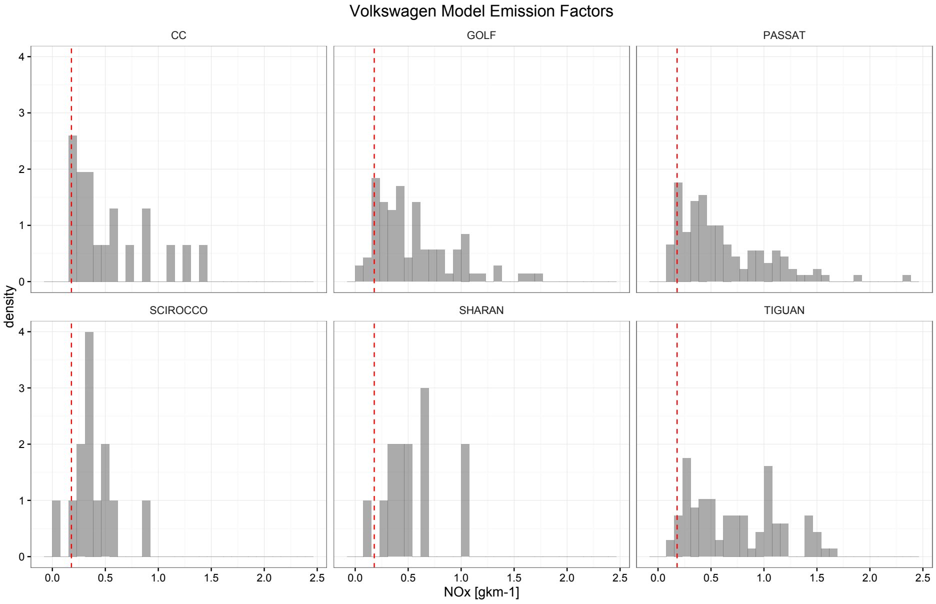

Figure 7.11: NO_X emission factors for Volkswagen models

Marque and Model	Location (a) [gkm^{-1}]	Scale (b)
Audi	0.35 ± 0.01	0.229 ± 0.008
Audi A1	0.43 ± 0.09	0.27 ± 0.07
Audi A3	0.39 ± 0.03	0.22 ± 0.02
Audi A4	0.32 ± 0.02	0.23 ± 0.02
Audi A5	0.36 ± 0.03	0.24 ± 0.03
Audi A6	0.36 ± 0.03	0.23 ± 0.03
Audi Q3	0.39 ± 0.02	0.21 ± 0.02
Audi Q5	0.29 ± 0.02	0.22 ± 0.02
Audi TT	0.37 ± 0.06	0.23 ± 0.05
Volkswagen	0.44 ± 0.02	0.30 ± 0.01
VW CC	0.37 ± 0.06	0.26 ± 0.05
VW Golf	0.39 ± 0.03	0.26 ± 0.02
VW Passat	0.43 ± 0.03	0.29 ± 0.02
VW Scirocco	0.31 ± 0.05	0.17 ± 0.03
VW Sharan	0.43 ± 0.06	0.21 ± 0.05
VW Tiguan	0.54 ± 0.04	0.33 ± 0.03

Table 7.5: Location and scale parameters for Gumbel distribution fits including standard error

The higher kerb weight compared to the Golf, Passat and Scirocco of this vehicle model may also be responsible for this however the Sharan is a similar kerb weight but smaller sample size may be limiting the visibility of this pattern.

All the different parameters that might be influencing the total emissions distribution of the Volkswagen Group vehicle fleet fitted with the EA189 engine have been analysed. It has been shown that vehicle models and marques do not vary significantly across all the range of 2.0L EA189 engine equipped vehicles. As such it is reasonable to recombine these vehicles into one subset for further analysis. It has been shown that the peak of the VWG EA189 engine emission occurs at $0.386gkm^{-1}$. The legislated type approval limit value is $0.180gkm^{-1}$. Volkswagen group vehicles are therefore exhibiting a modal emission of 2.14 times the EU legislated limit value with extreme values being more than ten times higher in some cases.

These results demonstrate clearly that the VWG vehicles are indeed running at levels considerably higher than those required to pass the laboratory based type approval test. This is not at all surprising given that these vehicles have been fitted with defeat devices to allow this exact behaviour. The magnitude of the increase is not perhaps as high as some might have thought prior to this analysis. The most extreme cases, although very infrequent are still of concern. The Q-Q analysis presented in Figure 7.12 shows that these vehicles generally behave well within the extreme value distribution and hence the use of shape and scale parameters to describe the fleet is valid. The causes of these more extreme values is less likely to be due to malfunctioning or other external factors but is more likely to be part of the natural distribution of values associated with vehicles operating in real driving environments.

7.5.4 Performance of Other Vehicles Relative to EA189 Engines

NO_X emissions from diesel vehicles has long been understood as a problem that affects the whole fleet, not just Volkswagen Group. One of the greatest strengths of the RSD methodology is that it allows a fair comparison across vehicle marque and model. It would be unfair to only look at Volkswagen vehicles in this way. Given the volume of vehicle measurements available from this database simple tests can be performed to demonstrate where VWG rank against other manufacturers in terms of on-road vehicle emissions. Further analysis can

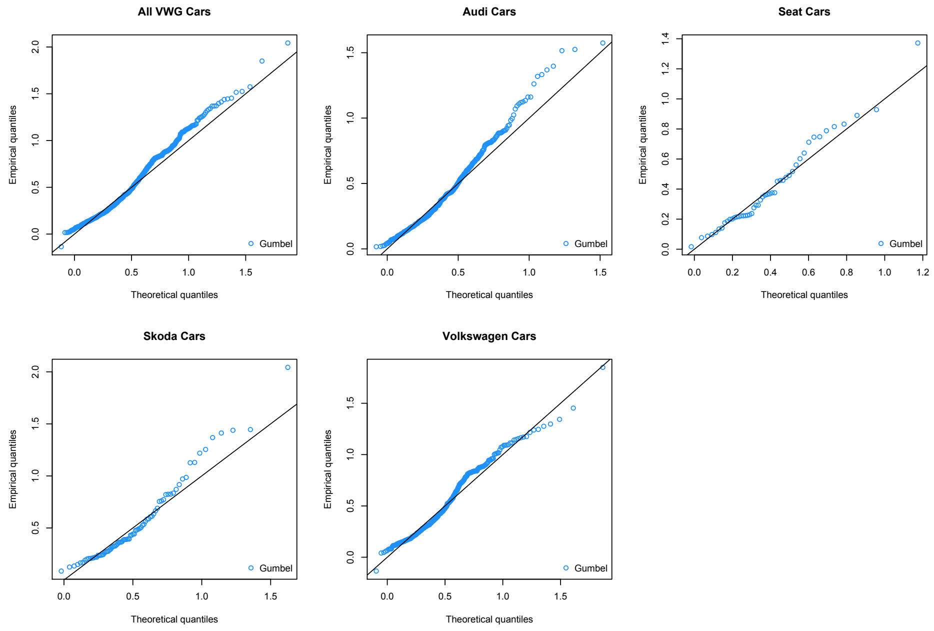

Figure 7.12: QQ plots of NO_X emission factors for VWG marques

help to understand which vehicles are most efficient and which are the least.

The rest of the vehicles observed in the Aberdeen study were cut down to those which were Euro 5 diesel cars with an engine capacity greater than $1.9L$ and less than $2.1L$. This data cut is the most representative of the VWG vehicles that were studied previously whilst allowing some variation in engine capacity between different manufacturers. Any vehicles that were badged as a VWG marque were also removed to prevent cross contamination between the samples. The remaining vehicles had an emission factor calculated in the same way as the VWG vehicles did with the f_{NO2} and CO_2 factors being the same. Information regarding the fractional NO_2 of different vehicle marques has been presented in Figure 18 (page 36) of Carslaw & Rhys-Tyler (2013) however the manufacturer identities are anonymised and unavailable so not considered at this stage. The variation between manufacturers, whilst not as small as for Euro 4 vehicles presented in the same figure, are similar so without further information the only option for this analysis that remains is to treat all Euro 5 diesel cars as having the same fractional NO_2 content. In the future analysis where fractional NO_2 for each marque is available would be needed to confirm or refute this assumption.

A minimum sample size of 20 vehicles was required per manufacturer for further analysis. The following marques were removed from analysis because their sample size was too small: Alfa Romeo, Chevrolet, Fiat, Mercedes, Nissan, Opel, Renault, Saab, Ssangyong, Subaru and Suzuki. The emission factors are calculated and are presented in Figure 7.13. As with Figures 7.11, 7.9 and 7.10 relating to VWG engines the Euro 5 diesel limit value is indicated as a red dashed line. The fitted distribution functions are indicated in blue for VWG and in green for the rest of the vehicles analysed. The position parameter for the whole set of non-EA189 engines was estimated by fitting a Gumbel distribution to the data set. The position parameter was estimated as 0.48 ± 0.01 compared to the same parameter for the VWG subsection which is estimated as 0.30 ± 0.01.

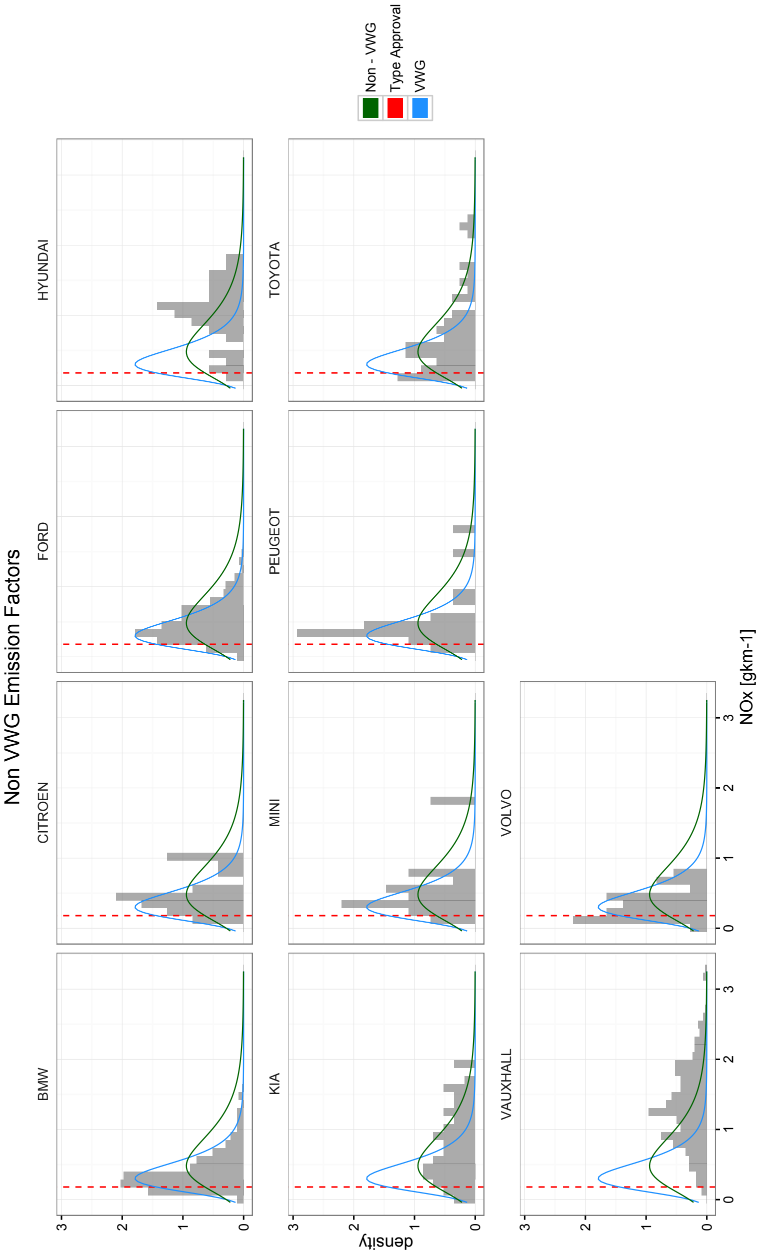

Figure 7.13: Distributions of NO_X emission factors for non-VWG marques

When the observations of the rest of the fleet are compared to VWG the immediate conclusion is that VWG are not noticeably worse than any other manufacturer but considerably better than some. When compared to the rest of the fleet as a whole the VWG vehicles have a lower peak or modal number and a more compact distribution resulting in less influence by the high emitting tail. There are only a small number of off-model vehicles (Section 5.4.2) in the Euro 5 diesel passenger car category however VWG are largely on-model when considered as a whole fleet. This is counterintuitive if the assumption that VWG are the only manufacturer using a defeat device or employing steps to reduce its NO_X emissions that are only effective over controlled laboratory based drive cycles is to believed. From these observations it could be hypothesised that other manufacturers are running similar or alternative defeat devices on their vehicles with a manufacturer of immediate interest being Vauxhall. A Gumbel distribution is fitted to each of the vehicle marques shown in Figure 7.13 so that a quantitative comparison can be made. The fitted parameters are shown in Table 7.6

Q-Q plots for all of the marques calculated in this subsection are presented in Figure 7.14. For the vast majority of vehicles marques the Gumbel distribution is a fair description of the fleet. The distribution analysis shows that for the most part the vehicles behave in a way that is consistent with the Gumbel distribution for all non VWG models. The a parameter for the non VWG models is higher than for the VWG distribution suggesting that the Volkswagen Group vehicles are performing better on-road than the other manufacturers. When compared to isolated vehicle marques the VWG vehicles are statistically out-performing 8 of the 10 manufacturers analysed. The exceptions being BMW and Volvo. This is surprising given that the use of the defeat device in VWG vehicles should mean it doesn't perform as well as those vehicles with effective and active emissions control systems.

Marque and Model	Location ($a[gkm^{-1}]$)	Scale (b
Non-VWG	0.47 ± 0.01	0.39 ± 0.01
BMW	0.28 ± 0.01	0.19 ± 0.01
Citroen	0.36 ± 0.05	0.20 ± 0.04
Ford	0.36 ± 0.01	0.21 ± 0.01
Hyundai	0.76 ± 0.08	0.43 ± 0.06
Kia	0.54 ± 0.06	0.42 ± 0.05
Mini	0.37 ± 0.05	0.25 ± 0.04
Peugeot	0.34 ± 0.05	0.22 ± 0.04
Toyota	0.46 ± 0.05	0.37 ± 0.04
Vauxhall	1.03 ± 0.03	0.52 ± 0.02
Volvo	0.23 ± 0.03	0.17 ± 0.02
VWG	0.30 ± 0.01	0.20 ± 0.01

Table 7.6: Location and scale parameters for Gumbel distribution fits for all major manufacturers

The inference from here is that either the use of emissions control devices does not impact the tailpipe emissions of the vehicles on the road or that other manufacturers are using similar systems to circumvent the emissions type approval process. Neither of these results reflect favourably on the auto industry.

The results of this analysis show that whilst EA189 engine vehicles are definitely emitting more NO_X than the type approval limit value they are as good as any other vehicle of a similar class to them on the road. More often than not the EA189 engine outperforms it's rivals when measured under real driving conditions. This is a positive result for VWG however the implications are concerning. Since no other manufacturer has been caught using a defeat device this result suggests that drive cycle beating is employed by multiple engine manufacturers.

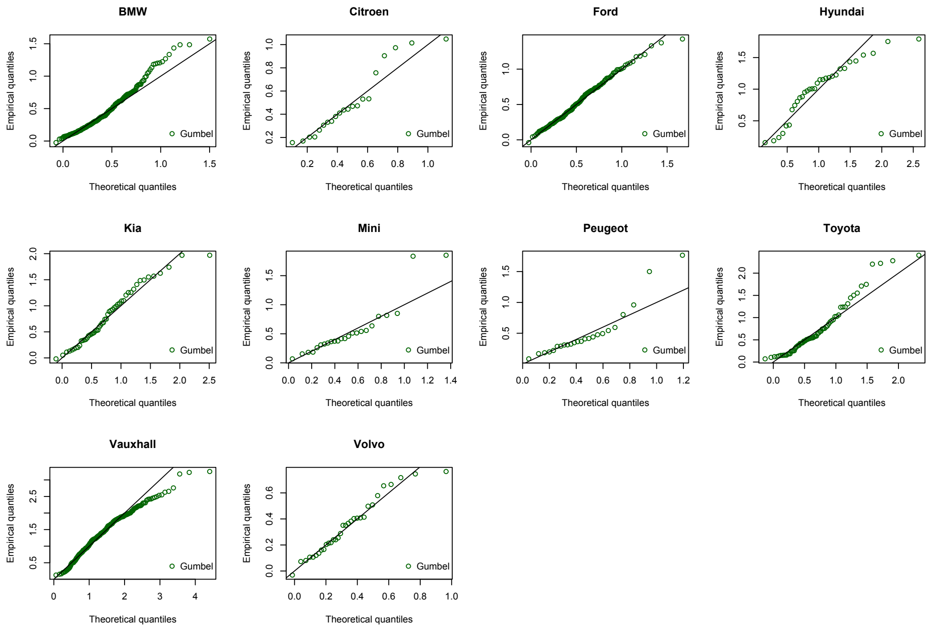

Figure 7.14: Q-Q plots for NO_X emission factors for non-VWG marques

A further result shown by this analysis is that passenger vehicle manufacturers as well as previously identified in light commercial vehicle manufacturers (Section 7.3) have very different solutions to reducing vehicle emissions with some being considerably more successful than others. The position parameter for emissions by Vauxhall vehicles for example is 7.4 times greater than the type approval limit or 3.5 times greater than the VWG position parameter. The vast majority of manufacturers manage to avoid emitting emissions at this rate and BMW, Ford and Volvo have managed to produce fleets of vehicles that emit at a low peak value and are also able to maintain control over their extreme emissions emitters.

7.5.5 Emissions of Down Sized Petrol Turbo Engines

There is some evidence that modern petrol powered passenger cars are emitting an increasingly high level of primary NO_2 (Carslaw & Rhys-Tyler, 2013). Observed f_{NO2} values for these vehicles has risen from a couple of percent to over 10% from 2005 onwards. A cause of this may be the vehicles fitted with downsized and turbo boosted petrol engines that have been introduced to reduce the total CO_2 emissions of the fleet (EC, 2014). A The hypothesis that downsized engines are responsible for this increase can be tested as one of the shorter research questions identified in Section1.3 using remotely sensed vehicle emission observations and vehicle type meta-data. Assessing this vehicle subset has proven to be challenging as it is not clear exactly where this technology has been integrated into the fleet and which manufacturers have decided to use alternative systems however educated guesses can be taken by subsetting based on aspiration (turbo or natural), fuel type and engine size and configuration.

Ford first introduced downsized engines in 2007 under the EcoBoost badge. A number of different engine configurations including a high and low power three cylinder 999 cubic centimetre (cc) engine alongside larger 4 and 6 cylinder varieties. In the Aberdeen data set 101 high power three cylinder measurements and 91 low power three cylinder engines measurements were taken along with 27 measurements of four-cylinder vehicles. These vehicles are subset from the rest of the fleet for separate analysis. The measured ratio of $NO : CO_2$ is compared to those vehicles which may or may not have GDI engines and are Euro 5 passenger cars. Two comparison sets are made comprising of vehicles with engine capacity $990cc - 1100cc$ and $1550cc - 1650cc$ to compare with the small and large Ford EcoBoost engines respectively. To best minimise cross contamination vehicles badged as Ford

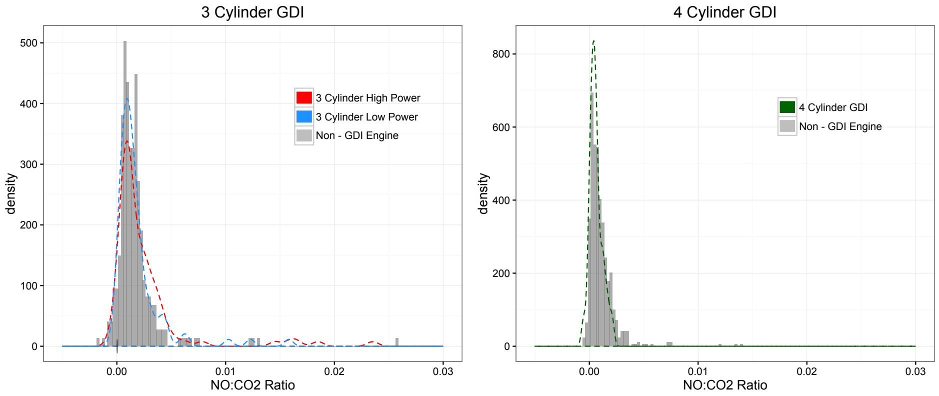

Figure 7.15: Distribution of downsized vehicles compared to the rest of the fleet

are all removed from the non-GDI sample set. The total NO_X emission factor is not calculated in this experiment as no f_{NO_2} information is available to distinguish different types of vehicles however NO is of primary concern as it is produced during the lean burn conditions tested. Use of emission factors calculated in the way previously described would, in this case, invalidate the result. If the GDI engines are causing an increase in NO due to their lean burning operation this will be observable without the need to calculate emission factors.

The emissions ratios are presented in Figure 7.15. The grey histogram shows the non-Ford vehicles and the frequency polygons show the GDI candidate vehicles. For the low and high power three cylinder engines there is no discernible difference between the Ford EcoBoost vehicles and the rest of the fleet for both the high and low variety. For the four cylinder vehicles there is evidence that the GDI fitted vehicles are performing slightly better than the rest of the fleet. As previously discussed in Section 7.5.4 Ford vehicles are typically one of the better manufacturers so this result is in line with those previously reported. The distribution of the $NO : CO_2$ ratio are fitted to a Gumbel distribution as shown in previous sections and the results are presented in Table 7.7.

Fleet Section	Location ($a \times 10^4$)	Scale ($b \times 10^4$)
Ford EcoBoost 3C High	14 ± 2	16 ± 1
Ford EcoBoost 3C Low	12 ± 1	13 ± 1
$1.0L$ Petrol Fleet	10.0 ± 0.7	11.1 ± 0.5
Ford EcoBoost 4C	3.0 ± 1.0	4.8 ± 0.7
$1.6L$ Petrol Fleet	6.2 ± 0.3	6.8 ± 0.2

Table 7.7: Location and scale parameters for Gumbel distribution fits of GDI vehicles

Analysis of the fitted parameters show that there is little statistically significant difference between the $1.0L$ fleet and the three cylinder Ford EcoBoost engines measured in Aberdeen. A two sample K-S test was performed on both the low and high powered 3 cylinder GDI vehicle sets against the $1.0L$ petrol fleet with $p_{low} = 0.91$ and $p_{high} = 0.01$ suggesting that there is some evidence of the high powered vehicles not coming from the same continuous distribution as the non GDI vehicles. The use of arbitrary cutoff points in p statistic analysis can be misleading (Wasserstein & Lazar, 2016) and the high powered vehicles appear to be largely similar to the $1.0L$ fleet. A two-sample K-S test was also applied to the 4 cylinder GDI vehicles with $p = 0.08$ confirming the null hypothesis that the two data sets fit the same underlying distribution. The rest of the fleet appears to be slightly more efficient at removing the NO from the exhaust gasses however the standard errors suggest that the two different engine configurations are comparable. This is a useful result from a modelling perspective as it means that the fleet does not need to be subsetted to account for this technology. The EcoBoost four cylinder engines outperform the rest of the fleet in a statistically meaningful way both in terms of the peak emissions and the spread of the more extreme values. The Q-Q plots for these distribution fit parameters are shown in Figure 7.16.

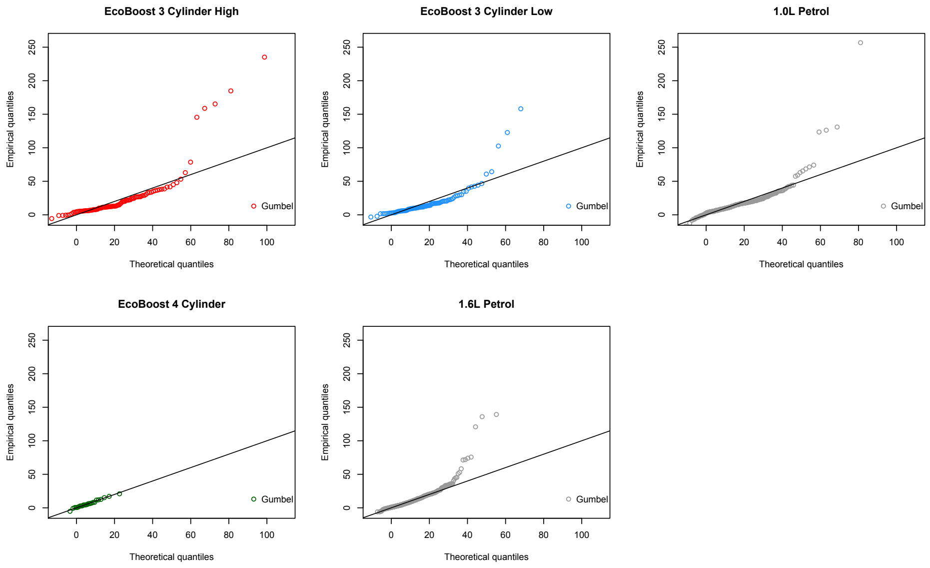

Figure 7.16: Q-Q plots for the data presented in Table 7.7

The Q-Q plots show that the data largely fit well to the Gumbel distribution with the four cylinder EcoBoost vehicles proving to be a particularly good fit. There are a disproportionately high number of three cylinder EcoBoost vehicles that exhibit off model behaviour as described in Section 5.4.2. For a sample size of 101 and 91 vehicles, the number of vehicles exhibiting this characteristic was 6 and 6 respectively. The number of vehicles exhibiting this characteristic in the rest of the fleet was 11 from a sample size of 294 or approximately three times more frequent in the GDI set. This is not observed in the 4 cylinder set with all the vehicles behaving on-model. When this is contrasted with the 1.6L normal vehicles there are a lot of off-model behaviour vehicles. The reason for this is unknown however if the EcoBoost 4 cylinder vehicles are consistently running without failure then the reason behind this should be a point of interest for future research.

The emissions of petrol powered cars are not currently of concern to policy makers trying to reduce the total NO_X emitted into urban environments as the vast majority of it still originates from diesel vehicles. Despite this if petrol powered vehicles continue to emit more and more NO_X in the future then these vehicles should come under scrutiny as well. There is no evidence that Ford Eco-boost vehicles are emitting any more NO than comparable vehicles with an unknown air-fuel ratio however this conclusion may be due to not knowing the leanness of the other vehicles. If the observed trend of increased NO_X emission from petrol cars continues it will be worth further investigation of emissions from vehicles running with different air-fuel ratios. As it stands Ford continue to emit at a level consistent with or better than the equivalent vehicles of different marques present in the rest of the fleet.

7.5.6 Conclusions and Discussion

This section has focussed on the 2.0 litre Euro 5 diesel engine and was motivated by the Volkswagen Group scandal. The key question to be answered was how do VWG vehicles perform in real driving environments and how does this compare to the rest of the fleet. This section has investigated the NO_X emissions of VWG vehicles and similar vehicles as they were measured in real driving environments in Aberdeen and have been assigned CO_2 values consistent with this driving environment. It has been shown that VWG marque vehicles with the EA189 engine of the $2.0L$ variety perform as well as or better than all comparable vehicle marques as a fleet. The VWG vehicles have been shown to emit as much or less than all the rest of the equivalent vehicles from all major marques. Given that VWG are the only manufacturer currently implicated in the defeat device scandal this result was not expected and suggests that drive cycle beating is present in all major manufacturers to some level. The RSD cannot tell if a defeat device is operating however by analysing a large number of vehicles and looking at how the fleet behaves as a whole rather than individual vehicles emissions the similarities in behaviours across marques is consistent with some sort of cycle beating taking place. It is also shown that there is significant difference in the effectiveness of the emission reduction systems installed in $2.0L$ diesel powered vehicles between manufacturers. Manufacturers such as Ford, Peugeot and BMW consistently outperform manufacturers such as Hyundai, Kia and especially Vauxhall.

A first attempt at analysing the impact of lean burn GDI technology has been completed. The results are currently inconclusive. Some evidence of higher NO_X emissions has been observed in some cases but not in others. It remains to be seen exactly how these vehicles will impact the fleet. It has also been shown that the lower engine capacity turbo aspirated vehicles are less efficient at reducing their NO_X emissions than the larger capacity petrol engines utilising the same technology. These engines have been introduced as a response to the requirement for manufacturers to reduce their total fleet CO_2 emissions and their NO_X emissions which have traditionally have been low for petrol cars may have been neglected in real driving conditions. The impact of these vehicles remains an open question but remote sensing devices have been shown to be capable of assessing it.

7.6 Conclusions and Discussion

In this chapter analysis has been performed using remote sensing device data on a range of real world problems. The impact of an ULEZ has been estimated, the emissions of light commercial vehicles have been analysed separately from the passenger car fleet, taxis have been compared to privately owned vehicles and the Volkswagen scandal has been analysed in a way only possible using remote sensing device data.

The estimates of the ULEZ impact on fleet emissions show clearly that the introduction of such schemes will produce reductions in NO_X in excess of anything that could be achieved if the fleet is allowed to turn over to Euro 6 naturally. Real driving environment measurements are not available for Euro 6b and Euro 6c compliant vehicles and given the stricter testing nature required to meet these levels there is still some scope for future reductions in NO_X to occur naturally. It is recommended that once Euro 6b / 6c vehicles have been observed in real driving conditions and have been parameterised in the same way as the current Euro 6 fleet these projections are re-visited. The projection is further limited in that it does not account for the NO_X emitted by LCVs and HCVs, busses or taxis nor does it take into account any reduction in vehicle usage or mode choice change after the introduction of an ULEZ. Such questions are beyond the scope of this thesis however the work presented here could be used as a platform from which future work could begin.

The ULEZ analysis examined two different scenarios for fleet change. A maximum spend and a minimum spend. Surprisingly but not counterintuitively the maximum spend did not produce as great a reduction in NO_X as the minimum spend achieved however they were both beyond what would be expected by 2025 in the NAEI fleet projections. The more effective minimum spend was caused by people moving from old diesels to old petrols which still emit less NO_X than the newest diesels. It has therefore been shown that whilst an ULEZ will produce noticeable effects on the NO_X emissions if it is used in conjunction with other push factors designed to increase the uptake of petrol or other low NO_X emitting vehicles then the reduction in NO_X can be further increased. Aberdeen has a fleet fraction of Euro 5 diesel vehicles higher than would be expected throughout the rest of Scotland and focusing on changing these vehicles to petrol, hybrid or electric vehicles as an addition to the ULEZ scheme could reduce the total NO_X emissions even more.

It has been shown that the light commercial vehicle fleet contributes a non-trivial amount of NO_X to the overall fleet emissions inventory and are unsurprisingly emitting higher than the legislated limit values. The impact of Euro 6 LCVs was not assessed as there were no vehicles meeting this criteria measured throughout the course of data collection. The NO_X emissions were calculated using two different methodologies, one using CO_2 factors measured in external studies and one using computer simulations and vehicle tracking performed on the streets of Aberdeen. The different methodologies agreed with each other to the accuracy of $\approx 1\%$. Attempts to model these vehicles in the same way that was achieved for the passenger car fleet have been confounded by the difference in effectiveness of NO_X emission reduction systems between different manufacturers. Given the significant differences in distributions observed and the time constraints on this thesis solutions to these problems have not been investigated however they represent an important and achievable area for future research.

The release of new LCVs into the fleet by different manufacturers has also been investigated. The results show that different manufacturers release their newest vehicles at different times. This further confounds the previously stated issues with modelling the LCVs as the fleet mix function is dependent on the manufacturer split as well. Further work must be done in this area to best understand how these vehicles are introduced into the fleet and when that is understood a robust attempt can be made to model them in the same way as passenger car vehicles.

The difference between private hire cars, hackney carriage style vehicles and the passenger car fleet as a whole has been completed. Through fitting the vehicles to the Gumbel distribution and the normal distribution it has been shown in both cases that private hire vehicles emit more NO_X than privately owned vehicles. Hackney carriage vehicles also emit higher NO_X than a representative subset of the privately owned fleet. The taxi fleet represents between $5-10\%$ of the representative fleet and are responsible for 50% more NO_X than their private fleet counterparts. The large number of taxis present suggests that there is some room for further reduction of total fleet NO_X by considering these vehicles. The temporal distribution of taxis may be important in reducing NO_X emissions at times of peak roadside NO_X concentrations.

The criticism of VWG go far beyond the simple levels of NO_X their vehicles emit. Consumers are feeling betrayed or lied to with the blatant level of cheating that VWG had attempted to get away with in an area where the significant metrics related to health concerns are well known (Section 2.2.2). The research presented in this section has been shown that VWG are by no means the worst but are in fact one of the better manufacturers for producing lower emission diesel passenger cars. The reasons for this are unclear as the data collected during remote sensing surveys cannot assess the state of the emission control system on vehicles measured. It may be hypothesised that given the allowance for vehicles to switch off their systems under certain ambient conditions the majority of vehicles are simply running without any emissions countermeasures active however further work must be done using a combination of remote sensing and additional methodologies to properly understand this. Given the severe financial toll and the loss of consumer good-will that VWG have suffered in the wake of this scandal there are lessons to be learned by all manufacturers.

This result yet again demonstrates that consistency of results in the laboratory has little to no bearing on consistency of vehicles in real driving environments. The case for real driving emissions tests to form part of the type approval process is reenforced. This section and Section 7.3 have raised some scientifically interesting but concerning details about the difference between different manufacturers and their ability or willingness to eliminate NO_X emissions from their vehicles. At the end of the lifetime of the Euro 5 legislation a convergence of results that might be expected has not been observed. This is a concern for the stability of emissions of Euro 6 vehicles across the fleet. As yet not enough Euro 6 vehicles have been observed to test whether or not this will be the case with the new legislation but as new ultra-low emission zone schemes that rely on the consistency of Euro 6 vehicles as low emitters to function effectively the question remains very relevant.

In summary solutions to a number of real world problems have been proposed based on data collected using the remote sensing methodology. Through doing this a number of interesting and topical questions have been answered making use of the analytical tools developed earlier in the thesis. Each of these individual topics has policy implications further applications that can be developed. These are discussed in Chapter 8.

Chapter 8

Summary of Conclusions Discussions and Further Work

8.1 Summary

This chapter will summarise the work done throughout this thesis, assess it's impact and look to the future. Full conclusions are presented within their respective chapters. With vehicle emissions and air quality topics currently making headlines around the world there should be a wide scope of opportunity for these projects to be expanded.

Four large scale remote sensing surveys were completed over three years. These were located in Sheffield, Cambridge, Aberdeen and a series of controlled experiments off the public highway. Additional smaller on-road studies were completed in Leeds. From these studies a number of key research questions have been answered.

The first objective stated Section 1.3 was to improve the ability of the RSD4600 to measure total NO_X emissions. The commercial RSD4600 used by the University of Leeds has been shown to be in good agreement for NO measurements with the prototype FEAT system used by the University of Denver. From this result it has been possible to use fractional NO_2 values measured by the FEAT system in conjunction with the NO measurements measured by the RSD4600 to expand the measurement capabilities of the RSD4600 to estimate total NO_X emissions using real data from vehicles. This is something that was not possible before. Previously total NO_X emissions of vehicles was estimated using roadside modelling methodology and the measurement driven NO_X method derived in this thesis has been shown to be an improvement.

The data collected using the RSD4600 can now be used to now estimate total NO_X emissions. Combining this data with the CO_2 emissions of the vehicle has been achieved by building on the work of others in assessing the true CO_2 emissions of vehicles. This has been done using a range of methods including instantaneous vehicle emissions models (Section 2.5.3) and large Europe-wide surveys investigating the true fuel consumption of vehicles (Section 3.3.2). A more accurate picture has been created with regards to the total NO_X emitted by vehicles on UK roads than was available prior to this research being completed.

The second objective stated in Section 1.3 was to improve the mathematical framework for describing and analysing emissions of vehicle fleets. Extreme value distribution mathematics, specifically the Gumbel distribution, has been shown to be a good description of the distribution of vehicle emissions of individual classes of passenger cars (Section 5.2.1) and a range of real world tests have been performed (Section 5.3). Using this method of description it has been possible to more realistically describe the fleet behaviour and relevant information can now be extracted from the data. Rather than simply expressing emissions in terms of the normal distribution parameterised by a mean and a standard deviation it has been shown that the Gumbel distribution parameterised by the location and scale parameters gives more information about the distribution of the vehicle emissions across the whole fleet. The use of the location and scale parameters provide information more relevant to vehicle emissions as the location parameter relates to the peak or modal emission and the scale parameter contains information about the impact of the most highly emitting vehicles. These physically relevant parameters, especially the modal number, are more useful for describing changes and differences in real world terms than the rate and shape parameters of the gamma distribution.

Using this new description it has been shown that the fleet of petrol powered vehicles and new diesel powered vehicles feature a non-trivial fraction of off-model vehicles. These vehicles that do not follow the Gumbel distribution have also been studied and a number of hypotheses for their existence have been proposed and tested using this new framework. The hypothesis that they are related to cold start emissions has been analysed and no strong connection has been made. Further work that is targeted at cold start only vehicles is required to further the knowledge in this area. The hypothesis that the off-model vehicles are caused by failed or otherwise non-functioning catalytic converter systems has been investigated by modelling the vehicles as Euro 0 for petrol and Euro 5 for diesel. This solution has been very successful at replicating the observed distribution of vehicles in the petrol fleet and it has been asserted that the cause of the high levels of NO_X that these vehicles emit is related to the operating condition of the catalyst. The same methodology has been successful in the diesel fleet however the success is not as obvious as for the petrol fleet because the effect is not as pronounced. A larger sample of Euro 6 diesel vehicles is required to fully understand this behaviour. Explaining this behaviour in terms of failed catalysts has proven to be the most effective methodology developed in this thesis.

8. SUMMARY OF CONCLUSIONS DISCUSSIONS AND FURTHER WORK

One motivation behind creating this model was to assess the effectiveness of the introduction of Euro 6 emission limit values. For the first time a large number of Euro 6 passenger vehicles have been observed in real driving conditions and the effectiveness of the new legislation has been able to be assessed. The result suggests that whilst they are better than Euro 5 they are still falling short of the emission factors required for homologation when they are observed in real driving environments even considering conformity factors. A relatively small Euro 6 passenger car sample size has been observed compared to other vehicle subsets. The reduction in total NO_X that these vehicles are able to achieve over their lifetime remains an open question and worthy of further investigation. The introduction of stricter emissions standards for the Euro 6b and Euro 6c vehicles is a question that can be answered as these vehicles are introduced into the fleet from 2017 and the RSD methodology will be the quickest way to test their emissions real world driving conditions. These results should be fed back into models for assessment as soon as the observations are available. To make this observation possible it is recommended that the Department for Transport ensure the subclass of Euro 6 that a vehicle is part of is part of the vehicle registration database.

Beyond being a useful tool for describing the vehicle emissions parameterising the distribution of emissions using the Gumbel distribution can be extended to provide a more realistic mathematical description that is suitable for modelling. Whilst the model presented in this thesis is far from complete, for example no treatment of LCVs or HCVs are presented, it nonetheless shows the potential that this method has for making predictions on policy outcomes. This is currently an understudied area. Microsimulation models coupled with instantaneous vehicle emission models are capable of calculating total emission factors over whole networks but they have a number of limitations. Firstly the vehicles do not represent the vehicle fleet as chassis dynamometer tests cannot be performed on all vehicles. The model coupled with remote sensing device measurements taken in the network that is to be modelled improves the efficacy of these measurements greatly. PHEM style modelling does not take into account the very highly emitting vehicles that contribute a significant amount of pollution to the total inventory. The model presented in this thesis has a functionality that allows assessment of this contribution to be made.

The model presented in this thesis is relatively simple to use if the data is in the right format and if valid CO_2 measurements are available. The model can be ran on a desktop computer using open source software in a short time period making it widely accessible. As such this tool will provide policy makers with an additional method to help them with the decision making process. Intervention policies can be invasive and unpopular so it is important to use the best tools available ensuring that any such policy is undertaken with the best possible information underpinning it.

The final objective stated in Section 1.3 is to apply the tools developed and the lessons learned to a number of real world problems. These questions have answers that are directly related to policy decisions. To this end a number of different vehicle subsets have been analysed on a level playing field for the first time. The impact of taxis compared to privately owned vehicles has been investigated and it has been shown that taxis emit more than private vehicles of comparable specification.

A number of research questions have been framed in the context of an ultra-low emission zone in Aberdeen. Policy ideas have been compared to a do-minimum scenario based on simulations created in Section6.4 and potential scenarios for an ULEZ described in Section 7.2. It has been shown that modernising the fleet to new diesel powered vehicles is unlikely to reduce the total NO_X emitted by the fleet as a whole. It has also been shown that a move from newer diesel vehicles to older petrol vehicles would reduce the total NO_X emitted by the fleet. It is hypothesised that a move towards a petrol powered fleet that contains a high number of hybrid vehicles would be the best solution to NO_X emitted by the taxi fleet. A wide range of further fleet distribution scenarios can be tested using this methodology and remains an area for further work.

The emissions of LCVs has also been investigated with a view to introducing them into the model developed throughout the thesis. Significant variance in emission distribution from make and marque has been observed. It remains to be seen whether or not this will present a significant obstacle in modelling. Further tests are required to determine the best way to proceed with modelling this subsection of the fleet.

The differences between different make and model of diesel passenger cars have been investigated in a topical investigation based on the Volkswagen Group scandal. It has been shown that VWG vehicles measured by the RSD emit more NO_X than their type approval limit and are often significantly higher. It has also been shown that VWG vehicles are not the highest emitting make and manufacturer with a range of different results being observed across all major manufacturers. No evidence currently exists to show that other manufacturers are using defeat devices to pass the type approval tests however given the magnitude and distribution of emission factors from non-VWG vehicles it is hypothesised that introduction of defeat devices or other methods of drive-cycle cheating have been employed by some or all manufacturers of Euro 5 diesel vehicles.

8.2 Further Work

One key aim of this thesis that was not achieved was to understand the effectiveness of Euro 6 legislation on the NO_X output of light commercial vehicles. An attempt was made to measure these vehicles in the Aberdeen study. The attempt was unsuccessful as no Euro 6 light commercial vehicles were measured. It is hypothesised that this is because the vehicles were yet to enter the fleet measured by the RSD. Light commercial vehicle emissions remain of significant interest to those who are interested in understanding and modelling pollution in urban environments and the current inability to measure them in real driving environments significantly reduces the ability of modellers to predict the future levels of pollution. As time progresses it is expected and required by law that these new Euro 6 vehicles will be introduced into the fleet so future remote sensing studies will have direct access to this information. Using the tools provided in this thesis and additional data collected on these vehicles an accurate assessment of the impact of the Euro 6 legislation on this section of the fleet will be possible and relatively easily achievable in small numbers without the need for a large measurement campaign. A small sample size could be measured in Leeds after a short survey lasting only one or two days or a larger set would likely be observed in an extended observation project. More robust statistics would be achievable for a larger sample size and increasing the measurement time to one or two weeks would naturally increase the accuracy of any statement made about Euro 6 light commercial vehicles.

There is some evidence that the fractional NO_2 of newer petrol powered vehicles is increasing. A possible cause for this may be the lean burning technologies, gasoline direct injection systems for example, in modern petrol cars aimed at reducing CO_2 emissions (Carslaw & Rhys-Tyler (2013), Figure 13, page 31). Traditionally petrol powered vehicles have emitted very little NO_2 as there is no need to introduce an oxidation catalyst to the system. Use of the FEAT(Section 2.6.5) remote sensing device has identified this issue and preliminary research has shown that there is an increase in NO_X emission by GDI engines in some cases but not in others. Further work in this area is required to better understand the contribution of these engines relative to the rest of the fleet. Whilst not currently identified as a major concern the trends identified in this work and previous surveys suggests that if left unresolved the emissions from these vehicles may present a problem in the future. Extended remote sensing of these vehicles under both controlled driving and real driving conditions along with concurrent PEMS data will be required to fully assess this question.

The modelling methodology that predicts NO_X for various fleet subsections can be extended to predict natural fleet changes and the impact this would have on the environment. A projection based on the NAEI fleet change prediction has been made however the model used for fleet turnover cannot fully take into account the changes in the fleet. Models are currently being developed at the University of Leeds (amongst others) that provide a considerably more robust method for handling changes in fleet distribution however they are not complete. Once these fleet models are completed the outputs can be used as inputs for the vehicle emissions model. The results generated from a model such as this would provide a more realistic control or do nothing scenario model for which all intervention programs such as the London Ultra-Low Emission Zone could be tested against than the simple case presented in this thesis. The comparison of the control to the currently achievable step change model used in Section **??** give policy decision makers more information and they can make better decisions about the most effective way to reduce pollution in urban environments. If the effects of the London ULEZ would occur naturally over 5 years with a better fleet model there is significantly less motivation to implement a costly and potentially unpopular piece of legislation however if it could be shown in a robust way that the ULEZ introduction was the only way it was possible to reduce total pollution below a threshold level then the politician may be more motivated to pursue the legislation. The impact of the change to WLTP and PEMS based testing for Euro 6b and Euro 6c should not be underestimated and an RSD based survey of these vehicles should be undertaken as soon as possible.

The RSD reports measurements for HC, CO and PM in addition to the NO measurements used in this thesis. Modelling the emissions of these pollutants has not been attempted as NO_X is currently of concern to policy makers. The methods developed in this thesis can be applied to the other pollutants too. It is suggested that the modelling of PM emissions is undertaken as a priority as the emissions from these vehicles is also of high importance.

Whilst constructing the model it became apparent that there were a small number of vehicles, often a few percent of a subsection, that were producing significantly higher emissions than the rest of the population. These vehicles were off-model even given the ability of the extreme value distribution to account for high emitting vehicles. These vehicles were often emitting NO_X with orders of magnitude greater than the mode of the distribution. It has been shown through modelling that these vehicles contribute a non-trivial amount of emission to the total inventory and identifying and fixing these vehicles alone represents an opportunity to reduce the overall pollution by up to 6% in some cases. This is a significant amount of pollution that can be removed from urban environments with a small amount of effort. The remote sensing device can identify these vehicles through post processing but it is unable to identify the cause of the problem. A number of causes have been suggested with modifications, faults and very poor optimisation being among potential candidates explored however the RSD cannot validate any of these hypotheses directly. Further steps taken in conjunction with other vehicle emissions assessment measured could be used to identify the causes of these super highly emitting vehicles.

The Weibull distribution is identified as an extreme value distribution useful for describing the failure rate of items. It is hypothesised that this function could be used to predict the rate of failure of catalyst systems in vehicles in the future. The current form of the model assumes that the failure rate remains constant. To confirm or refute this hypothesis for Euro 6 diesels would require further measurements to be made over a timespan of years. The number of Euro 6 vehicles that are subject to failure could then be modelled as a function of time and if it matched the Weibull distribution the fit parameters could be fine tuned to better predict the impact of these vehicles.

8.3 Final Thoughts

Vehicles emitting pollution into urban environments is an issue that has not yet been solved and significant challenges remain in solving the problem. Nonetheless the problem may yet be solvable. The more data that can be acquired the better models can be made and the greater our understanding will become. The wealth of information currently available tells us that this problem will not be solved by one method alone and requires a holistic approach encompassing information from static laboratories, portable emissions laboratories, modelling and remote sensing. This thesis has shown that remote sensing has a significant role to play in the solving of this problem and has developed some tools that will be useful for answering future research questions. The sheer volume of data collected during a remote sensing survey can appear disordered compared to some of the more well established methods however it is just as valuable and fills significant gaps in the research that the other methods are unable to fill. No other method can take a snapshot of the whole fleet's emission profile and attempts to do so can lead to errors in assumptions. Attempts can be made to simulate real driving but the remote sensing device is the only method that observes a large number of real drivers driving real cars on real roads under real driving conditions and the value of this cannot be understated.

With the introduction of new legislation during the course of this PhD and the subsequent uptake through various fleet sectors only now expected to become visible now is a very exciting time to be studying in-situ vehicle emissions. Remote sensing will be the first methodology that is able to directly observe the effects of these legislative steps. The motivation to understand these problems before the London ULEZ is introduced is high. There may be commercial applications for this research as more cities want to expand their understanding of their own air quality problems and search for solutions to fix them.

Throughout this thesis the shortcomings of the RSD4600, it's inability to measure NO_2, it's relatively high error compared to other methodologies and it's lack of ability to cover the whole network have been addressed and largely solutions have been provided through combination studies with other instruments and methodologies. Further to this the remote sensing device has been shown as a tool that can be useful in making predictions that other methodologies are unable to do. It would be impossible for chassis dynamometer tests to accurately represent the whole fleet of Aberdeen, Sheffield or Cambridge but the remote sensing device has achieved this. Emissions factors calculated using remote sensing device data have also been shown to be necessary for predicting the impact of significant pieces of legislation. By using up to date fleet information and emissions measured in situ the remote sensing device data modelled mathematically represents a bespoke tool that can be used to convince policy makers willing to invest in the data collection or extrapolate from other data sets if preferred.

The remote sensing methodology is a data first methodology. The style of science that uses data first and works backwards to find the relationship is alien to the methodologies used by scientists since Newton. Once the vehicle has left the highly controlled laboratory conditions everything changes. The traditional hypothesis, model, measure, conclusion method is not always suitable for the highly chaotic environment that measuring vehicle emissions can be. Google through the `http://cloud.google.com` service amongst others have been the pioneers in data driven research by collecting as much data as they can and figuring out how to use it later (Mayer-Schönberger & Cukier, 2013). Businesses such as Spotify, Coca-Cola, Philips and London Heathrow are all taking advantage of the big data revolution. The remote sensing device is the only tool currently available that can reach the whole fleet and measure it. There will always be a place for controlled laboratory studies, in fact their contribution is just as valid as any remote sensing study, however given the tools presented in this thesis the remote sensing device must now be seen as being on the same level of the more traditional vehicle emission measurement methodologies for affecting real change in the pollution levels in urban environments.

References

Abbott, J. & Stedman, J. (2005). Primary nitrogen dioxide emissions from road traffic: analysis of monitoring data. *AEA Technology, National Environmental Technology Centre. Report AEAT-1925*. 83, 143

Ahn, K., Rakha, H., Trani, A. & Van Aerde, M. (2002). Estimating vehicle fuel consumption and emissions based on instantaneous speed and acceleration levels. *Journal of transportation engineering*, **128**, 182–190. 53

Aksoy, H. (2000). Use of gamma distribution in hydrological analysis. *Turkish Journal of Engineering and Environmental Sciences*, **24**, 419–428. 157

Anderson, R.F., ALLENSWORTH, D.C. & DEGROOT, W.J. (1967). Myocardial toxicity from carbon monoxide poisoning. *Annals of internal medicine*, **67**, 1172–1182. 20

Anderson, T.W. & Darling, D.A. (1952). Asymptotic theory of certain" goodness of fit" criteria based on stochastic processes. *The annals of mathematical statistics*, 193–212. 134

Andersson, J., May, J., Favre, C., Bosteels, D., de Vries, S., Heaney, M., Keenan, M. & Mansell, J. (2014). On-road and chassis dynamometer evaluations of emissions from two euro 6 diesel vehicles. *SAE International Journal of Fuels and Lubricants*, **7**, 919–934. 46, 74

André, M. (2004). The artemis european driving cycles for measuring car pollutant emissions. *Science of the total Environment*, **334**, 73–84. 38, 39

Andrews, G.E. & Ahamed, F.M. (1999). The composition of spark ignition engine steady state particulate emissions. Tech. rep., SAE Technical Paper. 36

Andrews, G.E., Zhu, G., Li, H., Simpson, A., Wylie, J.A., Bell, M. & Tate, J. (2004). The effect of ambient temperature on cold start urban traffic emissions for a real world si car. 38

Andrews, G.E., Li, H., Wylie, J.A., Zhu, G., Bell, M. & Tate, J. (2005). Influence of ambient temperature on cold-start emissions for a euro 1 si car using in-vehicle emissions measurement in an urban traffic jam test cycle. Tech. rep., SAE Technical Paper. 38

Archer, G. (2015). Mind the gap 2015: Closing the chasm between test and real-world car co2 emissions. 87

Aris, R.M., Christian, D., Hearne, P.Q., Kerr, K., Finkbeiner, W.E. & Balmes, J.R. (1993). Ozone-induced airway inflammation in human subjects as determined by airway lavage and biopsy. *American Review of Respiratory Disease*, **148**, 1363–1372. 18

Armor, J.N. (1992). Environmental catalysis. *Applied Catalysis B: Environmental*, **1**, 221–256. 23

Artelt, S., Kock, H., König, H., Levsen, K. & Rosner, G. (1999). Engine dynamometer experiments: platinum emissions from differently aged three-way catalytic converters. *Atmospheric Environment*, **33**, 3559–3567. 23

ASHBAUGH, L. & LAWSON, D. (1991). A comparison of emissions from mobile sources using random roadside surveys conducted in 1985, 1987, and 1989. 63

ASHBAUGH, L.L., LAWSON, D.R., BISHOP, G.A., GUENTHER, P.L., STEDMAN, D.H., STEPHENS, R.D., GROBLICKI, P.J., PARIKH, J.S., JOHNSON, B.J. & HAUNG, S. (1992). On-road remote sensing of carbon monoxide and hydrocarbon emissions during several vehicle operating conditions. *Environmental Source Controls, Phoenix, AZ*. 60

ATKINSON, R.W., BARRATT, B., ARMSTRONG, B., ANDERSON, H., BEEVERS, S.D., MUDWAY, I.S., GREEN, D., DERWENT, R.G., WILKINSON, P., TONNE, C. *et al.* (2009). The impact of the congestion charging scheme on ambient air pollution concentrations in london. *Atmospheric Environment*, **43**, 5493–5500. 48

BACCARELLI, A., MARTINELLI, I., ZANOBETTI, A., GRILLO, P., HOU, L.F., BERTAZZI, P.A., MANNUCCI, P.M. & SCHWARTZ, J. (2008). Exposure to particulate air pollution and risk of deep vein thrombosis. *Archives of Internal Medicine*, **168**, 920–927. 15

BAHREINI, R., MIDDLEBROOK, A., GOUW, J.D., WARNEKE, C., TRAINER, M., BROCK, C., STARK, H., BROWN, S., DUBE, W., GILMAN, J. *et al.* (2012). Gasoline emissions dominate over diesel in formation of secondary organic aerosol mass. *Geophysical Research Letters*, **39**. 13

BAN-WEISS, G.A., MCLAUGHLIN, J.P., HARLEY, R.A., LUNDEN, M.M., KIRCHSTETTER, T.W., KEAN, A.J., STRAWA, A.W., STEVENSON, E.D. & KENDALL, G.R. (2008). Long-term changes in emissions of nitrogen oxides and particulate matter from on-road gasoline and diesel vehicles. *Atmospheric Environment*, **42**, 220–232. 50

Becker, E.R. & Watson, R.J. (1998). Future trends in automotive emission control. Tech. rep., SAE Technical Paper. 23, 24

Becker, K., Lörzer, J., Kurtenbach, R., Wiesen, P., Jensen, T. & Wallington, T. (1999). Nitrous oxide (n2o) emissions from vehicles. *Environmental science & technology*, **33**, 4134–4139. 83

Beevers, S.D. & Carslaw, D.C. (2005). The impact of congestion charging on vehicle emissions in london. *Atmospheric Environment*, **39**, 1–5. 48

Bensalah, Y. *et al.* (2000). *Steps in applying extreme value theory to finance: a review*. Bank of Canada. 154

Bielaczyc, P., Szczotka, A. & Woodburn, J. (2014). Cold start emissions of spark-ignition engines at low ambient temperatures as an air quality risk. *Archives of Environmental Protection*, **40**, 87–100. 186

Bierlaire, M., Bolduc, D. & McFadden, D. (2008). The estimation of generalized extreme value models from choice-based samples. *Transportation Research Part B: Methodological*, **42**, 381–394. 154

Bishop, G. & Stedman, D. (1996). Measuring the emissions of passing cars. *Accounts of Chemical Research*, **29**, 489–495. 59, 60, 63

Bishop, G., Holubowitch, N. & Stedman, D. (2009). Remote measurements of on-road emissions from heavy-duty diesel vehicles in california. *Year*, **1**, 2008. 6, 79, 137, 139, 141

Bishop, G.A. (2007). On-road remote sensing of automobile emissions in the denver area: Year 5, january 2005. 66

Bishop, G.A. & Stedman, D.H. (1990). On-road carbon monoxide emission measurement comparisons for the 1988-1989 colorado oxy-fuels program. *Environmental Science & Technology*, **24**, 843–847. 59

Bishop, G.A. & Stedman, D.H. (2008). A decade of on-road emissions measurements. *Environmental Science & Technology*, **42**, 1651–1656. 62

Bishop, G.A., Burgard, D.A. & Stedman, D.H. (????). On-road remote sensing of automobile emissions in the la brea area: Year 3, october 2003. 66

Bishop, G.A., Starkey, J.R., Ihlenfeldt, A., Williams, W.J. & Stedman, D.H. (1989). Ir long-path photometry: a remote sensing tool for automobile emissions. *Analytical Chemistry*, **61**, 671A–677A. 59

Bishop, G.A., Stedman, D.H., De La Garza Castro, J. & Dávalos, F.J. (1997). On-road remote sensing of vehicle emissions in mexico. *Environmental science & technology*, **31**, 3505–3510. 37

Bishop, G.A., Morris, J.A., Stedman, D.H., Cohen, L.H., Countess, R.J., Countess, S.J., Maly, P. & Scherer, S. (2001). The effects of altitude on heavy-duty diesel truck on-road emissions. *Environmental science & technology*, **35**, 1574–1578. 57

Bishop, G.A., Peddle, A.M., Stedman, D.H. & Zhan, T. (2010). On-road emission measurements of reactive nitrogen compounds from three california cities. *Environmental science & technology*, **44**, 3616–3620. 62

Bishop, G.A., Stedman, D.H., Burgard, D.A. & Atkinson, O. (2016). High-mileage light-duty fleet vehicle emissions: Their potentially overlooked importance. *Environmental science & technology*. 157

Boam, D., Finlay, I., Biddulph, T., Ma, T., Lee, R., Richardson, S., Bloomfleld, J., Green, J., Wallace, S., Woods, W. *et al.* (1994). The sources of unburnt hydrocarbon emissions from spark ignition engines during cold starts and warm-up. *Proceedings of the Institution of Mechanical Engineers, Part D: Journal of Automobile Engineering*, **208**, 1–11. 20

Boland, P.J. (2007). *Statistical and probabilistic methods in actuarial science*. CRC Press. 157

Borken-Kleefeld, J. & Chen, Y. (2015). New emission deterioration rates for gasoline cars–results from long-term measurements. *Atmospheric Environment*, **101**, 58–64. 66, 166

Brandt, E.P., Wang, Y. & Grizzle, J.W. (2000). Dynamic modeling of a three-way catalyst for si engine exhaust emission control. *IEEE Transactions on control systems technology*, **8**, 767–776. 23

Breslow, L. & Goldsmith, J. (1958). Health effects of air pollution. *American Journal of Public Health and the Nations Health*, **48**, 913–917. 2

Brook, R.D., Rajagopalan, S., Pope, C.A., Brook, J.R., Bhatnagar, A., Diez-Roux, A.V., Holguin, F., Hong, Y., Luepker, R.V., Mittleman, M.A. *et al.* (2010). Particulate matter air pollution and cardiovascular disease an update to the scientific statement from the american heart association. *Circulation*, **121**, 2331–2378. 15

Brousse, B., Vidal, B., Ponticq, X., Goupil, G. & Alary, R. (2005). Pollution dispersion at an urban motorway tunnel portal: comparison of the small-scale predictive study with the actual conditions measured on the site. *Atmospheric Environment*, **39**, 2459–2473. 50

Browne, M., Allen, J., Nemoto, T. & Visser, J. (2010). Light goods vehicles in urban areas. *Procedia-Social and Behavioral Sciences*, **2**, 5911–5919. 251

Brunekreef, B. (1997). Air pollution and life expectancy: is there a relation? *Occupational and environmental medicine*, **54**, 781. 2

Brunekreef, B. & Holgate, S.T. (2002). Air pollution and health. *The lancet*, **360**, 1233–1242. 2

Burgard, D.A., Dalton, T.R., Bishop, G.A., Starkey, J.R. & Stedman, D.H. (2006). Nitrogen dioxide, sulfur dioxide, and ammonia detector for remote sensing of vehicle emissions. *Review of scientific instruments*, **77**, 014101. 62

Burnett, R.T., Smith-Doiron, M., Stieb, D., Raizenne, M.E., Brook, J.R., Dales, R.E., Leech, J.A., Cakmak, S. & Krewski, D. (2001). Association between ozone and hospitalization for acute respiratory diseases in children less than 2 years of age. *American journal of epidemiology*, **153**, 444–452. 18

Butt, J. (2012). *Activation, deactivation, and poisoning of catalysts*. Elsevier. 26

Butt, J. & Petersen, E. (1988). Poisoning of catalystsacademic press. *New York*. 26, 27

Carol, L., Newman, N. & Mann, G. (1989). High temperature deactivation of three-way catalyst. Tech. rep., SAE Technical Paper. 25

Carslaw, D. & Rhys-Tyler, G. (2013). Remote sensing of no2 exhaust emissions from road vehicles. *A report to the City of London Corporation and London Borough of Ealing, Version 16th July*. 3, 48, 49, 58, 65, 83, 84, 143, 164, 169, 224, 253, 276, 281, 287, 303

Carslaw, D., Beevers, S., Westmoreland, E., Williams, M., Tate, J., Murrells, T., Stedman, J., Li, Y., Grice, S., Kent, A. *et al.* (2011a). Trends in nox and no2 emissions and ambient measurements in the uk. *Defra, London*. 1, 34, 48, 66

Carslaw, D.C. (2005). Evidence of an increasing no 2/nox emissions ratio from road traffic emissions. *Atmospheric Environment*, **39**, 4793–4802. 3, 30, 49

Carslaw, D.C. & Beevers, S.D. (2004). New directions: Should road vehicle emissions legislation consider primary no 2? *Atmospheric Environment*, **38**, 1233–1234. 62

Carslaw, D.C., Beevers, S.D. & Fuller, G. (2001). An empirical approach for the prediction of annual mean nitrogen dioxide concentrations in london. *Atmospheric Environment*, **35**, 1505–1515. 35

Carslaw, D.C., Beevers, S.D., Tate, J.E., Westmoreland, E.J. & Williams, M.L. (2011b). Recent evidence concerning higher no x emissions from passenger cars and light duty vehicles. *Atmospheric Environment*, **45**, 7053–7063. iv, 3, 24, 34, 58

Carslaw, D.C., Williams, M.L., Tate, J.E. & Beevers, S.D. (2013). The importance of high vehicle power for passenger car emissions. *Atmospheric Environment*, **68**, 8–16. 58, 124, 170, 174, 176

Carslaw, D.C., Priestman, M., Williams, M.L., Stewart, G.B. & Beevers, S.D. (2015). Performance of optimised scr retrofit buses under urban driving and controlled conditions. *Atmospheric Environment*, **105**, 70–77. 32, 140

Carson, B., Ellis III, H., Beall, C. & Baker, L. (1981). Ammonia health effects. final report. Tech. rep., Midwest Research Inst., Kansas City, MO (USA); Environmental Protection Agency, Ann Arbor, MI (USA). Emission Control Technology Div. 32

Chan, T. & Ning, Z. (2005). On-road remote sensing of diesel vehicle emissions measurement and emission factors estimation in hong kong. *Atmospheric Environment*, **39**, 6843–6856. 61

Chan, T., Ning, Z., Leung, C., Cheung, C., Hung, W. & Dong, G. (2004). On-road remote sensing of petrol vehicle emissions measurement and emission factors estimation in hong kong. *Atmospheric Environment*, **38**, 2055–2066. 61

Chang, T., Modzelewski, S., Norbeck, J. & Pierson, W. (1981). Tunnel air quality and vehicle emissions. *Atmospheric Environment (1967)*, **15**, 1011–1016. 50

Chen, Y. & Borken-Kleefeld, J. (2014). Real-driving emissions from cars and light commercial vehicles–results from 13 years remote sensing at zurich/ch. *Atmospheric Environment*, **88**, 157–164. 38, 39, 119, 120

Choudhary, A. & Gokhale, S. (2016). Urban real-world driving traffic emissions during interruption and congestion. *Transportation Research Part D: Transport and Environment*, **43**, 59–70. 21

Cloke, J., Cox, J., Hickman, A., Ellis, S., Ingrey, M. & Buchan, K. (2000). A low emission zone for london. *TRL REPORT 431*. 48

CMAQ (2002). *The Congestion Mitigation and Air Quality Improvement Program: Assessing 10 Years of Experience*, vol. 264. Transportation Research Board. 21

Coles, S., Bawa, J., Trenner, L. & Dorazio, P. (2001). *An introduction to statistical modeling of extreme values*, vol. 208. Springer. 156

Colvile, R., Hutchinson, E., Mindell, J. & Warren, R. (2001). The transport sector as a source of air pollution. *Atmospheric environment*, **35**, 1537–1565. 1, 13, 19, 20

COMEAP (2010). The mortality effects of long-term exposure to particulate air pollution in the united kingdom. 14

COMEAP (2015). Statement on the evidence for the effects of nitrogen dioxide on health. 1, 17

Dagel, J.F. (1998). *Diesel engine and fuel system repair*. Prentice Hall. 32

Daham, B., Andrews, G.E., Li, H., Partridge, M., Bell, M.C. & Tate, J. (2005a). Quantitying the effects of traffic calming on emissions using on-road measurement. 53

Daham, B., Andrews, G.E., Li, H., Rosario, B., Bell, M., Tate, J. & Ropkins, K. (2005b). Application of a portable ftir for measuring on-road emissions. 52

Daly, A. & Bierlaire, M. (2006). A general and operational representation of generalised extreme value models. *Transportation Research Part B: Methodological*, **40**, 285–305. 154

Danaiah, P., Kumar, R. & Kumar, V. (2012). Lean combustion technology for internal combustion engines: a review. *Science and Technology*, **2**, 47–50. 28

Daniel, J.I. & Bekka, K. (2000). The environmental impact of highway congestion pricing. *Journal of Urban Economics*, **47**, 180–215. 47

Dardiotis, C., Martini, G., Marotta, A. & Manfredi, U. (2013). Low-temperature cold-start gaseous emissions of late technology passenger cars. *Applied energy*, **111**, 468–478. 186

De Nevers, N. (1995). Air pollution control engineering. mcgrw-hill. *Inc., New York*. 2

Defra (2011). What are the causes of air pollution. 17, 18

DeFRA (2013). Air quality: economic analysis. 1

Delignette-Muller, M.L. & Dutang, C. (2015). fitdistrplus: An R package for fitting distributions. *Journal of Statistical Software*, **64**, 1–34. 134, 154, 160, 167, 277

Delmas, R., Serca, D. & Jambert, C. (1997). Global inventory of nox sources. *Nutrient cycling in agroecosystems*, **48**, 51–60. 19

Demuynck, J., Bosteels, D., De Paepe, M., Favre, C., May, J. & Verhelst, S. (2012). Recommendations for the new wltp cycle based on an analysis of vehicle emission measurements on nedc and cadc. *Energy Policy*, **49**, 234–242. 40

DfT (2010). Light goods vehicle co2 emissions study: Final report. 254

DfT (2015). National travel survey: England 2014. 263

DieselNet (2005). Selective catalytic reduction. 32

EA (2013). *DPF Mysteries*. `http://emissionsanalytics.com/dpf-regeneration-mysteries/`, accessed 3 May 2016. 34, 263

Easterly, C. (1994). Ozone health effects. 16

Eastwood, P. (2000). *Critical topics in exhaust gas aftertreatment*. 36

EC (1991). Council directive 91/441/eec. 43

EC (1994). Council directive 94/12/ec. 43

EC (1996). Council directive 96/69/ec. 43

EC (1999a). European union euro iii standard directive 1999/96/ec. 1, 3, 43

EC (1999b). European union euro iv standard directive 2002/80/ec. 1, 3, 36, 43

EC (1999c). European union euro iv standard directive 98/69/ec. 1, 3, 43

EC (2001). Eu directive 2001/116/ec. 3, 36, 37

EC (2007a). Council directive 715/2007/ec. 43

EC (2007b). Regulation (ec) no 715/2007 of the european parliament and of the council of 20 june 2007 on type approval of motor vehicles with respect to emissions from light passenger and commercial vehicles (euro 5 and euro 6) and on access to vehicle repair and maintenance information. 1, 3, 36, 43, 188

EC (2008). Eu directive 2008/50/ec. 3, 35

EC (2014). Regulation (eu) no 333/2014 of the european parliament and of the council of 11 march 2014 amending regulation (ec) no 443/2009 to define the modalities for reaching the 2020 target to reduce co 2 emissions from new passenger cars. 287

EC (2015). European commission - press release commission welcomes member states agreement on robust testing of air pollution emissions by cars. 46

EEA (2008). Sources of air pollution. 19

EINSTEIN, A. (1905). Zur electrodynamik bewegter. *Korper.Ann Phys*, **17**, 132. 72

EKSTRÖM, M., SJÖDIN, Å. & ANDREASSON, K. (2004). Evaluation of the copert iii emission model with on-road optical remote sensing measurements. *Atmospheric Environment*, **38**, 6631–6641. 66

EL-SHAWARBY, I., RAKHA, H. & AHN, K. (2005). Impact of catalytic converter on on-road vehicle emissions. 52, 53

Ellison, R.B., Greaves, S.P. & Hensher, D.A. (2013). Five years of londons low emission zone: Effects on vehicle fleet composition and air quality. *Transportation Research Part D: Transport and Environment*, **23**, 25–33. 48, 214, 242

EPA (1999). Nitrogen oxides (nox), why and how they are controlled. 19

Epling, W.S., Campbell, L.E., Yezerets, A., Currier, N.W. & Parks, J.E. (2004). Overview of the fundamental reactions and degradation mechanisms of nox storage/reduction catalysts. *Catalysis Reviews*, **46**, 163–245. 30, 31

Erdos, P. & Lehner, J. (1941). The distribution of the number of summands in the partitions of a positive integer. *Duke Math. J*, **8**, 335–345. 156

ESP (2005). *RSD 4600 Operator's Manual Edition 3.0*. 132, 135

Farrauto, R.J. & Heck, R.M. (1999). Catalytic converters: state of the art and perspectives. *Catalysis Today*, **51**, 351–360. 22, 24

Farrauto, R.J. & Voss, K.E. (1996). Monolithic diesel oxidation catalysts. *Applied Catalysis B: Environmental*, **10**, 29–51. 29

Favre, C., Bosteels, D. & May, J. (2013). Exhaust emissions from european market-available passenger cars evaluated on various drive cycles. Tech. rep., SAE Technical Paper. 74

Fonken, L., Xu, X., Weil, Z.M., Chen, G., Sun, Q., Rajagopalan, S. & Nelson, R.J. (2011). Air pollution impairs cognition, provokes depressive-like behaviors and alters hippocampal cytokine expression and morphology. *Molecular psychiatry*, **16**, 987–995. 2

FORASTIERE, F., STAFOGGIA, M., PICCIOTTO, S., BELLANDER, T., D'IPPOLITI, D., LANKI, T., VON KLOT, S., NYBERG, F., PAATERO, P., PETERS, A. *et al.* (2005). A case-crossover analysis of out-of-hospital coronary deaths and air pollution in rome, italy. *American journal of respiratory and critical care medicine*, **172**, 1549–1555. 15

FORZATTI, P. & LIETTI, L. (1999). Catalyst deactivation. *Catalysis today*, **52**, 165–181. 26

FRANCO, V., SÁNCHEZ, F.P., GERMAN, J. & MOCK, P. (2014). Real-world exhaust emissions from modern diesel cars. *communications*, **49**, 847129–102. 46, 220

FRÉCHET, M. (1928). Sur la loi de probabilité de l'écart maximum. In *Annales de la societe Polonaise de Mathematique*, [sn]. 156

FREY, H.C., ROUPHAIL, N.M., UNAL, A. & COLYAR, J. (2001). Emissions reduction through better traffic management: An empirical evaluation based upon on-road measurements. Tech. rep. 53

GATTO, N.M., HENDERSON, V.W., HODIS, H.N., JOHN, J.A.S., LURMANN, F., CHEN, J.C. & MACK, W.J. (2014). Components of air pollution and cognitive function in middle-aged and older adults in los angeles. *Neurotoxicology*, **40**, 1–7. 2

GAUDERMAN, W.J., AVOL, E., GILLILAND, F., VORA, H., THOMAS, D., BERHANE, K., MCCONNELL, R., KUENZLI, N., LURMANN, F., RAPPAPORT, E. *et al.* (2004). The effect of air pollution on lung development from 10 to 18 years of age. *New England Journal of Medicine*, **351**, 1057–1067. 2

GIESHOFF, J., SCHÄFER-SINDLINGER, A., SPURK, P., VAN DEN TILLAART, J. & GARR, G. (2000). Improved scr systems for heavy duty applications. Tech. rep., SAE Technical Paper. 31

GIRARD, J., CAVATAIO, G., SNOW, R. & LAMBERT, C. (2009). Combined fe-cu scr systems with optimized ammonia to nox ratio for diesel nox control. *SAE International Journal of Fuels and Lubricants*, **1**, 603–610. 31

GOLDSTEIN, M. (2008). Carbon monoxide poisoning. *Journal of Emergency Nursing*, **34**, 538–542. 19

GONZALEZ-VELASCO, J., ENTRENA, J., GONZÁLEZ-MARCOS, J., GUTIÉRREZ-ORTIZ, J. & GUTIÉRREZ-ORTIZ, M. (1994). Preparation, activity and durability of promoted platinum catalysts for automotive exhaust control. *Applied Catalysis B: Environmental*, **3**, 191–204. 23

GÖTSCHI, T., HEINRICH, J., SUNYER, J. & KÜNZLI, N. (2008). Long-term effects of ambient air pollution on lung function: a review. *Epidemiology*, **19**, 690–701. 17

GRICE, S., STEDMAN, J., KENT, A., HOBSON, M., NORRIS, J., ABBOTT, J. & COOKE, S. (2009). Recent trends and projections of primary no 2 emissions in europe. *Atmospheric Environment*, **43**, 2154–2167. 83, 84, 143

GUARDIAN (2016). London takes just one week to breach annual air pollution limits. 3

GUENTHER, P.L., STEDMAN, D.H., BISHOP, G.A., BEATON, S.P., BEAN, J.H. & QUINE, R.W. (1995). A hydrocarbon detector for the remote sensing of vehicle exhaust emissions. *Review of scientific instruments*, **66**, 3024–3029. 60

GUENTHER, P.L., STEDMAN, D.H. & LESKO, J.M. (1996). Prediction of im240 mass emissions using portable exhaust analyzers. *Journal of the Air & Waste Management Association*, **46**, 343–348. 52

Gumbel, E. (1935). Les valeurs extrêmes des distributions statistiques. In *Annales de l'institut Henri Poincaré*, vol. 5, 115–158. 156, 268

Gumbel, E.J. (1941). The return period of flood flows. *The annals of mathematical statistics*, **12**, 163–190. 156

Haass, F. & Fuess, H. (2005). Structural characterization of automotive catalysts. *Advanced Engineering Materials*, **7**, 899–913. 29

Hagman, R. & Amudsen, A. (2013). Exhaust emissions from vehicles with euro 6/vi technology. *Institute of transport economics, Norwegian Centre for Transport Research, TOI Report*, **1259**. 164

Hamra, G.B., Laden, F., Cohen, A.J., Raaschou-Nielsen, O., Brauer, M. & Loomis, D. (2015). Lung Cancer and Exposure to Nitrogen Dioxide and Traffic: A Systematic Review and Meta-Analysis. *Environ. Health Perspect.*, **123**, 1107–1112. 17

Hart, C., Koupal, J. & Giannelli, R. (2002). Epa's onboard analysis shootout: Overview and results. Tech. rep. 53

Hausberger, S. (2003). Simulation of real world vehicle exhaust emissions. *VKM-THD Mitteilungen, Heft/Vol 82, Verlag der Technischen Universitat Graz*. 55

Hausberger, S. (2010). Fuel consumption and emissions of modern passenger cars. *TU Graz*. 254

Hawker, P., Myers, N., H
thwohl, G., Vogel, H.T., Bates, B., Magnusson, L. & Bronnenberg, P. (1997). Experience with a new particulate trap technology in europe. Tech. rep., SAE Technical Paper. 33

HBEFA (2010). Handbook emission factors for road transport. *INFRAS*. 66

He, L.Y., Hu, M., Huang, X.F., Zhang, Y.H., Yu, B.D. & Liu, D.Q. (2006). Chemical characterization of fine particles from on-road vehicles in the wutong tunnel in shenzhen, china. *Chemosphere*, **62**, 1565–1573. 51

Hearder, J.N. (1874). Physiological Effects of Ozone. , **9**, 182–183. 17

Heck, R. & Farrauto, R. (1996). Automotive catalysts. *Automotive Engineering*, **104**. 26, 27

Heck, R.M. & Farrauto, R.J. (2001). Automobile exhaust catalysts. *Applied Catalysis A: General*, **221**, 443–457. 22

Heeb, N.V., Forss, A.M., Saxer, C.J. & Wilhelm, P. (2003). Methane, benzene and alkyl benzene cold start emission data of gasoline-driven passenger cars representing the vehicle technology of the last two decades. *Atmospheric Environment*, **37**, 5185–5195. 36

Heijne, V., Ligterink, N. & Stelwagen, U. (2016). 2016 emission factors for diesel euro-6 passenger cars, light commercial vehicles and euro-vi trucks. Tech. rep., TNO. 87, 150

Herreros, J., Gill, S., Lefort, I., Tsolakis, A., Millington, P. & Moss, E. (2014). Enhancing the low temperature oxidation performance over a pt and a pt–pd diesel oxidation catalyst. *Applied Catalysis B: Environmental*, **147**, 835–841. 29, 263

Heywood, J.B. (1988). *Internal combustion engine fundamentals*, vol. 930. Mcgraw-hill New York. 61

Holman, C., Harrison, R. & Querol, X. (2015). Review of the efficacy of low emission zones to improve urban air quality in european cities. *Atmospheric Environment*, **111**, 161–169. 47, 48

HONG, Y.C., LEE, J.T., KIM, H., HA, E.H., SCHWARTZ, J. & CHRISTIANI, D.C. (2002). Effects of air pollutants on acute stroke mortality. *Environmental Health Perspectives*, **110**, 187. 15

HORIBA (2004). Obs1300 instruction manual. 52

HORIBA (2006). Obs2000 instruction manual. 52

HROMÁDKO, J., MILER, P., HROMÁDKO, J., HÖNIG, V. & SCHWARZKOPF, M. (2010). The influence of three-way catalysts on harmful emission production. *Transportation Research Part D: Transport and Environment*, **15**, 103–107. 27

HU, Z. & HECK, R. (1995). High temperature ultra stable close-coupled catalysts. Tech. rep., SAE Technical Paper. 24

HUSSAIN, J., PALANIRADJA, K., ALAGUMURTHI, N. & MANIMARAN, R. (2012). Effect of exhaust gas recirculation (egr) on performance and emission characteristics of a three cylinder direct injection compression ignition engine. *Alexandria Engineering Journal*, **51**, 241–247. 16

IARC (2013). Outdoor air pollution a leading environmental cause of cancer deaths. 1

ICCT (2015). European vehicle market statistics pocketbook 2015/16. 251

JANSSEN, N.A., BRUNEKREEF, B., VAN VLIET, P., AARTS, F., MELIEFSTE, K., HARSSEMA, H. & FISCHER, P. (2003). The relationship between air pollution from heavy traffic and allergic sensitization, bronchial hyperresponsiveness, and respiratory symptoms in dutch schoolchildren. *Environmental health perspectives*, **111**, 1512. 2

Jimenez-Palacios, J.L. (1998). *Understanding and quantifying motor vehicle emissions with vehicle specific power and TILDAS remote sensing*. Ph.D. thesis, Massachusetts Institute of Technology. 38, 41, 131

Joumard, R., Philippe, F. & Vidon, R. (1999). Reliability of the current models of instantaneous pollutant emissions. *Science of the total environment*, **235**, 133–142. 56

Joumard, R., André, M., Vidon, R. & Tassel, P. (2003). Characterizing real unit emissions for light duty goods vehicles. *Atmospheric Environment*, **37**, 5217–5225. 251

Kadijk, G., Ligterink, N. & Spreen, J. (2015). On-road nox and co2 investigations of euro 5 light commercial vehicles. *TNO report*, R10192. 85, 87, 253

Kampa, M. & Castanas, E. (2008). Human health effects of air pollution. *Environmental pollution*, **151**, 362–367. 1

Kassakian, J.G., Wolf, H.C., Miller, J.M. & Hurton, C.J. (1996). Automotive electrical systems circa 2005. *IEEE Spectr.*, **33**, 22–27. 38

Katare, S. & Laing, P.M. (2006). A hybrid framework for modeling aftertreatment systems: A diesel oxidation catalyst application. Tech. rep., SAE Technical Paper. 30

Katare, S.R., Patterson, J.E. & Laing, P.M. (2007). Diesel aftertreatment modeling: A systems approach to no x control. *Industrial & engineering chemistry research*, **46**, 2445–2454. 28

Kihara, N., Tsukamoto, T., Matsumoto, K., Ishida, K., Kon, M. & Murase, T. (2000). Real-time on-board measurement of mass emission of nox, fuel consumption, road load, and engine output for diesel vehicles. Tech. rep., SAE Technical Paper. 52

Koebel, M., Elsener, M. & Kleemann, M. (2000). Urea-scr: a promising technique to reduce no x emissions from automotive diesel engines. *Catalysis today*, **59**, 335–345. 62

Koltsakis, G.C. & Stamatelos, A.M. (1997). Catalytic automotive exhaust aftertreatment. *Progress in Energy and Combustion Science*, **23**, 1–39. 25

König, H., Hertel, R., Koch, W. & Rosner, G. (1992). Determination of platinum emissions from a three-way catalyst-equipped gasoline engine. *Atmospheric Environment. Part A. General Topics*, **26**, 741–745. 23

Konstandopoulos, A.G., Kostoglou, M., Skaperdas, E., Papaioannou, E., Zarvalis, D. & Kladopoulou, E. (2000). Fundamental studies of diesel particulate filters: transient loading, regeneration and aging. *SAE paper*, 1016. 33

Kourbatov, A. (2013). Maximal gaps between prime k-tuples: a statistical approach. *Journal of Integer Sequences*, **16**, 3. 156

Kousoulidou, M., Ntziachristos, L., Franco, V., Gkeivanidis, S., Samaras, Z. & Dilara, P. (2010). Use of portable emissions measurement system (pems) for validation and development of passenger car emission factors. In *18th International Symposium Transport and Air Pollution*. 74

Krajzewicz, D., Wagner, P., Woelki, M. & Dippold, M. (2015). Driving patterns reducing pollutant emission at traffic lights. 56

Kraschl-Hirschmann, K., Zallinger, M., Luz, R., Fellendorf, M. & Hausberger, S. (2011). A method for emission estimation for microscopic traffic flow simulation. In *Integrated and Sustainable Transportation System (FISTS), 2011 IEEE Forum on*, 300–305, IEEE. 56

Krivoshto, I.N., Richards, J.R., Albertson, T.E. & Derlet, R.W. (2008). The toxicity of diesel exhaust: implications for primary care. *The Journal of the American Board of Family Medicine*, **21**, 55–62. 2, 20

Kubsh, J.E. (1994). Alternative ehc heating patterns and their impact on cold-start emissions performance. Tech. rep., SAE Technical Paper. 25

Kulkarni, N. & Grigg, J. (2008). Effect of air pollution on children. *Paediatrics and Child Health*, **18**, 238–243. 105

Kumarathasan, P., Blais, E., Saravanamuthu, A., Bielecki, A., Mukherjee, B., Bjarnason, S., Guénette, J., Goegan, P. & Vincent, R. (2015). Nitrative stress, oxidative stress and plasma endothelin levels after inhalation of particulate matter and ozone. *Particle and fibre toxicology*, **12**, 1. 18

Lafyatis, D.S., Ansell, G.P., Bennett, S.C., Frost, J.C., Millington, P.J., Rajaram, R.R., Walker, A.P. & Ballinger, T.H. (1998). Ambient temperature light-off for automobile emission control. *Applied Catalysis B: Environmental*, **18**, 123–135. 24

Laschober, C., Limbeck, A., Rendl, J. & Puxbaum, H. (2004). Particulate emissions from on-road vehicles in the kaisermühlen-tunnel (vienna, austria). *Atmospheric Environment*, **38**, 2187–2195. 50

Lashof, D.A. & Ahuja, D.R. (1990). Relative contributions of greenhouse gas emissions to global warming. 28

Lassi, U. (2003). *Deactivation correlations of Pd/Rh three-way catalysts designed for Euro IV Emission Limits: effect of ageing atmosphere, temperature and time*. Oulun yliopisto. 23, 24, 25, 26, 27

Lautenschlager, S., Wulf, H.C. & Pittelkow, M.R. (2007). Photoprotection. *The Lancet*, **370**, 528–537. 18

Lavoie, G.A., Heywood, J.B. & Keck, J.C. (1970). Experimental and theoretical study of nitric oxide formation in internal combustion engines. *Combustion science and technology*, **1**, 313–326. 16

Lawson, D.R., Groblicki, P.J., Stedman, D.H., Bishop, G.A. & Guenther, P.L. (1990). Emissions from lit-use motor vehicles in los angeles: A pilot study of remote sensing and the inspection and maintenance program. *Journal of the Air & Waste Management Association*, **40**, 1096–1105. 63, 65

LCC (2015). Hackney carriage and private hire vehicle licences issued in leeds. 263

Lightman, A.P. (1993). *Ancient light: our changing view of the universe*. Harvard University Press. 194

Ligterink, N.E. & Eijk, A.R. (2014). Update analysis of real-world fuel consumption of business passenger cars based on travelcard nederland fuelpass data. *TNO report TNO*, R11063. 85, 86

Liotta, L., Macaluso, A., Arena, G., Livi, M., Centi, G. & Deganello, G. (2002). A study of the behaviour of pt supported on ceo 2–zro 2/al 2 o 3–bao as no x storage–reduction catalyst for the treatment of lean burn engine emissions. *Catalysis today*, **75**, 439–449. 28

Liu, Z., Ge, Y., Tan, J., He, C., Shah, A.N., Ding, Y., Yu, L. & Zhao, W. (2012). Impacts of continuously regenerating trap and particle oxidation catalyst on the no 2 and particulate matter emissions emitted from diesel engine. *Journal of Environmental Sciences*, **24**, 624–631. 49

LONGO, L.D. (1977). The biological effects of carbon monoxide on the pregnant woman, fetus, and newborn infant. *American journal of obstetrics and gynecology*, **129**, 69–103. 19

LORUSSO, J., KAISER, E. & LAVOIE, G. (1981). Quench layer contribution to exhaust hydrocarbons from a spark-ignited engine. 60

LOX, E., ENGLER, B., JANSSEN, F., GARTEN, R., DALLA BETTA, R., SCHLATTER, J., AINBINDER, Z., MANZER, L., NAPPA, M., PARMON, V. *et al.* (1997). Environmental catalysis. *Handbook of heterogeneous catalysis*, 1569–1595. 22, 23, 27

LUND, A., SHIMADA, S. & SHIOTANI, M. (2011). *Principles and applications of ESR spectroscopy*. Springer Science & Business Media. 16

MA, C.J., TOHNO, S. & KASAHARA, M. (2004). A case study of the single and size-resolved particles in roadway tunnel in seoul, korea. *Atmospheric Environment*, **38**, 6673–6677. 50

MA, J. & FUKUDA, D. (2015). A hyperpath-based network generalized extreme-value model for route choice under uncertainties. *Transportation Research Part C: Emerging Technologies*, **59**, 19–31. 154

MAJEWSKI, W.A. (2001). Diesel particulate filters. *DieselNet.¡ https://www. dieselnet. com/tech/dpf. php*. 33

MAJEWSKI, W.A. & KHAIR, M.K. (2006). *Diesel emissions and their control*. Society of Automotive Engineers. 29, 30

MAROTTA, A., PAVLOVIC, J., CIUFFO, B., SERRA, S. & FONTARAS, G. (2015). Gaseous emissions from light-duty vehicles: Moving from nedc to the new wltp test procedure. *Environmental science & technology*, **49**, 8315–8322. 74

Martins, E.S., Stedinger, J.R. *et al.* (2000). Generalized maximum-likelihood generalized extreme-value quantile estimators for hydrologic data. *Water Resources Research*, **36**, 737–744. 154

Matsumoto, S., Ikeda, Y., Suzuki, H., Ogai, M. & Miyoshi, N. (2000). No x storage-reduction catalyst for automotive exhaust with improved tolerance against sulfur poisoning. *Applied Catalysis B: Environmental*, **25**, 115–124. 30, 31

Mayer, H. (1999). Air pollution in cities. *Atmospheric environment*, **33**, 4029–4037. 48

Mayer-Schönberger, V. & Cukier, K. (2013). *Big data: A revolution that will transform how we live, work, and think*. Houghton Mifflin Harcourt. 4, 5, 308

Mazzoleni, C., Moosmüller, H., Kuhns, H.D., Keislar, R.E., Barber, P.W., Nikolic, D., Nussbaum, N.J. & Watson, J.G. (2004). Correlation between automotive co, hc, no, and pm emission factors from on-road remote sensing: implications for inspection and maintenance programs. *Transportation Research Part D: Transport and Environment*, **9**, 477–496. 64

McConnell, R., Berhane, K., Gilliland, F., Molitor, J., Thomas, D., Lurmann, F., Avol, E., Gauderman, W.J. & Peters, J.M. (2003). Prospective study of air pollution and bronchitic symptoms in children with asthma. *American journal of respiratory and critical care medicine*, **168**, 790–797. 17

McDonald, P., Seljak, U., Burles, S., Schlegel, D.J., Weinberg, D.H., Cen, R., Shih, D., Schaye, J., Schneider, D.P., Bahcall, N.A. *et al.* (2006). The lyα forest power spectrum from the sloan digital sky survey. *The Astrophysical Journal Supplement Series*, **163**, 80. 73

McElroy, C. & Fogal, P. (2008). Ozone: From discovery to protection. *Atmosphere-ocean*, **46**, 1–13. 18

McGaughey, G.R., Desai, N.R., Allen, D.T., Seila, R.L., Lonneman, W.A., Fraser, M.P., Harley, R.A., Pollack, A.K., Ivy, J.M. & Price, J.H. (2004). Analysis of motor vehicle emissions in a houston tunnel during the texas air quality study 2000. *Atmospheric Environment*, **38**, 3363–3372. 50

McKee, D. (1993). *Tropospheric ozone: human health and agricultural impacts*. CRC Press. 18

Mdyusof, N.F.F., Ramli, N.A., Yahaya, A.S., Sansuddin, N., Ghazali, N.A. & Al Madhoun, W.A. (2011). Central fitting distributions and extreme value distributions for prediction of high pm 10 concentration. In *Multimedia Technology (ICMT), 2011 International Conference on*, 6263–6266, IEEE. 157

Miller, K.A., Siscovick, D.S., Sheppard, L., Shepherd, K., Sullivan, J.H., Anderson, G.L. & Kaufman, J.D. (2007). Long-term exposure to air pollution and incidence of cardiovascular events in women. *New England Journal of Medicine*, **356**, 447–458. 15

Morgan, C.R., Carlson, D.W. & Voltz, S.E. (1973). Thermal response and emission breakthrough of platinum monolithic catalytic converters. Tech. rep., SAE Technical Paper. 166

MoS (2015). Its not just vw: Official tester claims four more diesel car giants break toxic emissions limit. 242

Moulijn, J.A., Van Diepen, A. & Kapteijn, F. (2001). Catalyst deactivation: is it predictable?: What to do? *Applied Catalysis A: General*, **212**, 3–16. 26

NAEI (2012). Naei fleet projection. 194, 221, 242

Nakamura, H., Kihara, N., Adachi, M. & Ishida, K. (2002). Development of a wet-based ndir and its application to on-board emission measurement system. Tech. rep., SAE Technical Paper. 52

Narayanan, D.L., Saladi, R.N. & Fox, J.L. (2010). Review: Ultraviolet radiation and skin cancer. *International journal of dermatology*, **49**, 978–986. 18

North, R., Ochieng, W., Quddus, M., Noland, R. & Polak, J. (2004). Validation and characterization of a vehicle performance and emissions monitoring system. 52

Nova, I., Ciardelli, C., Tronconi, E., Chatterjee, D. & Bandl-Konrad, B. (2006). Nh 3–no/no 2 chemistry over v-based catalysts and its role in the mechanism of the fast scr reaction. *Catalysis Today*, **114**, 3–12. 31

Ntziachristos, L., Samaras, Z., Eggleston, S., Gorissen, N., Hassel, D., Hickman, A. *et al.* (2000). Copert iii. *Computer Programme to calculate emissions from road transport, methodology and emission factors (version 2.1), European Energy Agency (EEA), Copenhagen*. 66

Obama, B. (2012). Big data across the federal government. 5

O'Driscoll, R., ApSimon, H., Oxley, T. & Molden, N. (2016). Portable emissions measurement system (pems) data for euro 6 diesel cars and comparison with emissions modelling. 993 – 1005, The provisional proceedings of the 21st International Transport and Air Pollution Conference TAP 2016. 46, 53

Oh, S.H. & Eickel, C.C. (1988). Effects of cerium addition on co oxidation kinetics over alumina-supported rhodium catalysts. *Journal of Catalysis*, **112**, 543–555. 23

Olsson, L. & Fridell, E. (2002). The influence of pt oxide formation and pt dispersion on the reactions no 2 no+ 1/2 o 2 over pt/al 2 o 3 and pt/bao/al 2 o 3. *Journal of Catalysis*, **210**, 340–353. 30, 31

Olsson, L., Persson, H., Fridell, E., Skoglundh, M. & Andersson, B. (2001). A kinetic study of no oxidation and no x storage on pt/al2o3 and pt/bao/al2o3. *The Journal of Physical Chemistry B*, **105**, 6895–6906. 30

Ostop, R. & Ryder, L. (1989). Ute pass carbon monoxide emissions study. *City of Colorado Springs, Department of Utilities, Environmental Services Division*. 65

Pierson, W., Brachaczek, W., Hammerle, R., McKee, D. & Butler, J. (1978). Sulfate emissions from vehicles on the road. *Journal of the Air Pollution Control Association*, **28**, 123–132. 50

Pierson, W.R., Gertler, A.W. & Bradow, R.L. (1990). Comparison of the scaqs tunnel study with other on road vehicle emission data. *Journal of the Air & Waste Management Association*, **40**, 1495–1504. 50

Planck, M. (1900). zur theorie des gesetzes der energieverteilung im normalenspektrum. *Verh. Dtsch. Phys. Ges*, **2**, 237. 72

Pokharel, S.S., Bishop, G.A. & Stedman, D.H. (2000a). *On-road Remote Sensing of Automobile Emissions in the Chicago Area, Year 3*. Coordinating Research Council. 64

Pokharel, S.S., Bishop, G.A., Stedman, D.H. & Lindner, A.D.J. (2000b). Analysis of remote sensing data for development of i/m program evaluation protocols. *Ann Arbor*, **1001**, 48105. 64

Pokharel, S.S., Bishop, G.A. & Stedman, D.H. (2001). *On-road Remote Sensing of Automobile Emissions in the Los Angeles Area, Year 2*. Coordinating Research Council. 66

Poon, S.H., Rockinger, M. & Tawn, J. (2004). Extreme value dependence in financial markets: Diagnostics, models, and financial implications. *Review of financial studies*, **17**, 581–610. 154

Pope, C.A., Burnett, R.T., Thurston, G.D., Thun, M.J., Calle, E.E., Krewski, D. & Godleski, J.J. (2004). Cardiovascular mortality and long-term exposure to particulate air pollution epidemiological evidence of general pathophysiological pathways of disease. *Circulation*, **109**, 71–77. 15

Pope III, C.A. & Dockery, D.W. (2006). Health effects of fine particulate air pollution: lines that connect. *Journal of the air & waste management association*, **56**, 709–742. 15

Pope III, C.A., Burnett, R.T., Thun, M.J., Calle, E.E., Krewski, D., Ito, K. & Thurston, G.D. (2002). Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution. *Jama*, **287**, 1132–1141. 15

Popp, P.J., Bishop, G.A. & Stedman, D.H. (1999a). Development of a high-speed ultraviolet spectrometer for remote sensing of mobile source nitric oxide emissions. *Journal of the Air & Waste Management Association*, **49**, 1463–1468. 61

Popp, P.J., Bishop, G.A. & Stedman, D.H. (1999b). On-road remote sensing of automobile emissions in the chicago area: Year 2. *CRC Project E-23 Report*. 65, 66

Power, M.C., Weisskopf, M.G., Alexeeff, S.E., Coull, B.A., Spiro III, A. & Schwartz, J. (2011). Traffic-related air pollution and cognitive function in a cohort of older men. *Environmental health perspectives*, **119**, 682. 2

Prati, M.V., Meccariello, G., Della Ragione, L. & Costagliola, M.A. (2015). Real driving emissions of a light-duty vehicle in naples. influence of road grade. Tech. rep., SAE Technical Paper. 52

PRINETTO, F., GHIOTTI, G., NOVA, I., LIETTI, L., TRONCONI, E. & FORZATTI, P.V. (2001). Ft-ir and tpd investigation of the no x storage properties of bao/al2o3 and pt-bao/al2o3 catalysts. *The Journal of Physical Chemistry B*, **105**, 12732–12745. 30

R CORE TEAM (2015). *R: A Language and Environment for Statistical Computing*. R Foundation for Statistical Computing, Vienna, Austria. 134, 154, 160, 167

RAKHA, H., AHN, K. & TRANI, A. (2004). The vt-micro framework for modeling of hot stabilized light duty vehicle and truck emissions. *Transportation Research, Part D: Transport & Environment*, **9**, 49–74. 53

RAYSONI, A.U. & LI, W.W. (2009). Health impacts of traffic related air pollution. *Journal of Environmental Research And Development Vol*, **4**. 105

RILEY, R. (2016). *Developing Real Driving CO2 Emission Factors for Hybrid Cars Through on Road Testing and Microscale Modelling*. Ph.D. thesis, University of Leeds. 263, 271

ROGERSON, S. (2013). Developing the evidence base and business case for hybrid taxis. 263

ROKOSZ, M., CHEN, A., LOWE-MA, C., KUCHEROV, A., BENSON, D., PECK, M.P. & MCCABE, R. (2001). Characterization of phosphorus-poisoned automotive exhaust catalysts. *Applied Catalysis B: Environmental*, **33**, 205–215. 27

RÖPKE, S., SCHWEIMER, G. & STRAUSS, T. (1995). Nox formation in diesel engines for various fuels and intake gases. Tech. rep., SAE Technical Paper. 15

ROPKINS, K., BEEBE, J., LI, H., DAHAM, B., TATE, J., BELL, M. & ANDREWS, G. (2009). Real-world vehicle exhaust emissions monitoring: review and critical discussion. *Critical Reviews in Environmental Science and Technology*, **39**, 79–152. 37, 50, 52, 54, 58, 63, 64, 66

Rosin, P. (1933). The laws governing the fineness of powdered coal. *J. Inst. Fuel.*, **7**, 29–36. 156

Rouphail, N.M., Frey, H.C., Colyar, J.D. & Unal, A. (2001). Vehicle emissions and traffic measures: exploratory analysis of field observations at signalized arterials. In *80th Annual Meeting of the Transportation Research Board, Washington, DC*. 53

Rubin, M.B. (2001). The history of ozone. the schönbein period, 1839–1868. *Bull. Hist. Chem*, **26**, 40–56. 17

Russell, A. & Epling, W.S. (2011). Diesel oxidation catalysts. *Catalysis Reviews*, **53**, 337–423. 28, 31, 33

Samuel, S., Morrey, D., Fowkes, M., Taylor, D., Austin, L., Felstead, T. & Latham, S. (2004). The most significant vehicle operating parameter for real-world emission levels. Tech. rep., SAE Technical Paper. 38

Schrank, D.L. & Lomax, T.J. (2007). *The 2007 urban mobility report*. Texas Transportation Institute, Texas A & M University. 21

Schwartz, J. (1994). Air pollution and daily mortality: a review and meta analysis. *Environmental research*, **64**, 36–52. 3

Sensors, I. (2006). Semtech-g. on-board, in-use gasoline vehicle emissions an- alyzer. company web equipment brochure. 52

Sharma, P., Khare, M. & Chakrabarti, S. (1999). Application of extreme value theory for predicting violations of air quality standards for an urban road intersection. *Transportation Research Part D: Transport and Environment*, **4**, 201–216. 6, 157

Sideris, M. (1998). *Methods for monitoring and diagnosing the efficiency of catalytic converters: a patent-oriented survey*. Elsevier. 25

Sileghem, L., Bosteels, D., May, J., Favre, C. & Verhelst, S. (2014). Analysis of vehicle emission measurements on the new wltc, the {NEDC} and the {CADC}. *Transportation Research Part D: Transport and Environment*, **32**, 70 – 85. 40, 41

Singh, P. & Prasad, R. (2014). Catalytic abatement of cold-start vehicular co emissions. *Catalysis in Industry*, **6**, 122–127. 186

Stedman, D.H. & Bishop, G.A. (1990). An analysis of on-road remote sensing as a tool for automobile emissions control. *Illinois Dept. of Energy & Natural Resources*, 1–175. 65

Stedman, D.H., Bishop, G.A., Peterson, J.E. & Hosick, T. (1993). Provo pollution prevention program: A pilot study of the cost-effectiveness of an on-road vehicle emissions reduction program. *University of Denver/City of Provo, Utah*. 64, 65

Stedman, D.H., Bishop, G.A., Beaton, S.P., Peterson, J.E., Guenther, P.L., McVey, I.F. & Zhang, Y. (1994). *On-road remote sensing of CO and HC emissions in California*. The Division. 64, 65

Stemmler, K., Bugmann, S., Buchmann, B., Reimann, S. & Staehelin, J. (2005). Large decrease of voc emissions of switzerland's car fleet during the past decade: results from a highway tunnel study. *Atmospheric Environment*, **39**, 1009–1018. 51

Takada, Y., Miyazaki, T. & Iida, N. (2002). Study on local air pollution caused by nox from diesel freight vehicle. In *SAE 2002 World Congress*. 52

Takada, Y., Takada, N. & Iida, N. (2005). Transient nox characteristics of freight vehicles with egr system in real traffic conditions. *SAE transactions*, **114**, 1129–1138. 53

Tate, J. (2016). Vehicle emission measurement and analysis - aberdeen city council. 24, 56, 58, 65, 112, 137, 224, 246, 253, 276

Tate, J. & Connors, R. (2014). Mapping vehicle emissions through urban streets and intersections. 55

Tate, J.E. (2013a). Vehicle emission measurement and analysis - cambridge city council. 24, 41, 56, 58, 65, 107, 137

Tate, J.E. (2013b). Vehicle emission measurement and analysis - sheffield city council. 24, 41, 56, 58, 65, 101, 137

Taylor, K.C. (1993). Nitric oxide catalysis in automotive exhaust systems. *Catalysis ReviewsScience and Engineering*, **35**, 457–481. 23

Telegraph (2016). Diesel cars emit more pollution in cold weather, study finds. 4, 188

Thom, S.R. (2002). Hyperbaric-oxygen therapy for acute carbon monoxide poisoning. *New England Journal of Medicine*, **347**, 1105–1106. 19

Tietge, U., Zacharof, N., Mock, P., Franco, V., German, J., Bandivadekar, A., Ligterink, N. & Lambrecht, U. (2015). From laboratory to road: a 2015 update of official and real-world fuel consumption and co2 values for passenger cars in europe. *ICCT white paper*. 86, 164, 177

Times, N.Y. (2015). Volkswagen says 11 million cars worldwide are affected in diesel deception. 273

Turner, M.C., Krewski, D., Pope III, C.A., Chen, Y., Gapstur, S.M. & Thun, M.J. (2011). Long-term ambient fine particulate matter air pollution and lung cancer in a large cohort of never-smokers. *American journal of respiratory and critical care medicine*, **184**, 1374–1381. 15

Turrio-Baldassarri, L., Battistelli, C.L., Conti, L., Crebelli, R., De Berardis, B., Iamiceli, A.L., Gambino, M. & Iannaccone, S. (2004). Emission comparison of urban bus engine fueled with diesel oil and biodieselblend. *Science of the Total Environment*, **327**, 147–162. 36

Tutuianu, M., Marotta, A., Steven, H., Ericsson, E., Haniu, T., Ichikawa, N. & Ishii, H. (2013). Development of a world-wide worldwide harmonized light duty driving test cycle (wltc). Tech. rep., UNECE, Technical report. 4

Twigg, M.V. (2006). Roles of catalytic oxidation in control of vehicle exhaust emissions. *Catalysis Today*, **117**, 407–418. 23, 30, 33

VBOX (2008). Vbox ii lite 5hz gps data logger. 125

Vincent, R., Bjarnason, S.G., Adamson, I., Hedgecock, C., Kumarathasan, P., Guénette, J., Potvin, M., Goegan, P. & Bouthillier, L. (1997). Acute pulmonary toxicity of urban particulate matter and ozone. *The American journal of pathology*, **151**, 1563. 18

Vojtisek-Lom, M. & Allsop, J.E. (2001). Development of heavy-duty diesel portable, on-board mass exhaust emissions monitoring system with nox, co2 and qualitative pm capabilities. Tech. rep., SAE Technical Paper. 52

Walker, A. (2004). Controlling particulate emissions from diesel vehicles. *Topics in catalysis*, **28**, 165–170. 30

Walsh, P., Sagebiel, J., Lawson, D., Knapp, K. & Bishop, G. (1996). Comparison of auto emission measurement techniques. *Science of the total environment*, **189**, 175–180. 64

Wasserstein, R.L. & Lazar, N.A. (2016). The asa's statement on p-values: context, process, and purpose. *The American Statistician*. 289

Watanabe, T., Kawashima, K., Tagawa, Y., Tashiro, K., Anoda, H., Ichioka, K., Sumiya, S. & Zhang, G. (2007). New doc for light duty diesel dpf system. Tech. rep., SAE Technical Paper. 33

Weaver, C.S. & Balam-Almanza, M.V. (2001). Development of the ravemride-along vehicle emission measurement system for gaseous and particulate emissions. Tech. rep., SAE Technical Paper. 52

Weber, C., Hagman, R. & Amudsen, A. (2015). Emission from vehicles with euro 6/vi-technology results from the measurement programme emiroad 2014. *Institute of transport economics, Norwegian Centre for Transport Research, TOI Report*, **TI Report 1405/2015**. 164

Weilenmann, M., Soltic, P., Saxer, C., Forss, A.M. & Heeb, N. (2005). Regulated and nonregulated diesel and gasoline cold start emissions at different temperatures. *Atmospheric environment*, **39**, 2433–2441. 186

Weiss, M., Bonnel, P., Kühlwein, J., Provenza, A., Lambrecht, U., Alessandrini, S., Carriero, M., Colombo, R., Forni, F., Lanappe, G. *et al.* (2012). Will euro 6 reduce the no x emissions of new diesel cars?–insights from on-road tests with portable emissions measurement systems (pems). *Atmospheric Environment*, **62**, 657–665. 43, 46, 74

Wenzel, T., Singer, B.C. & Slott, R. (2001). Some issues in the statistical analysis of vehicle emissions. *Statistical Anaysis and Modeling of Automotive Emissions*, 1. 21

WHO (2006). Who air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide: global update 2005: summary of risk assessment. 1, 17, 30

WHO (2013). Review of evidence on health aspects of air pollution: Revihaap project: final technical report. 1

Wichmann, H.E. (2007). Diesel exhaust particles. *Inhalation toxicology*, **19**, 241–244. 14

Wilkins, E. (1954). Air pollution and the london fog of december 1952. *JR Sanit. Inst.;(United States)*, **74**. 3

Williams, M.L. & Carslaw, D.C. (2011). New directions: Science and policy–out of step on no x and no 2? *Atmospheric Environment*, **45**, 3911–3912. 1

Williamson, W., Perry, J., Goss, R., Gandhi, H. & Beason, R. (1984). Catalyst deactivation due to glaze formation from oil-derived phosphorus and zinc. Tech. rep., SAE Technical Paper. 27

Winkler, A., Dimopoulos, P., Hauert, R., Bach, C. & Aguirre, M. (2008). Catalytic activity and aging phenomena of three-way catalysts in a compressed natural gas/gasoline powered passenger car. *Applied Catalysis B: Environmental*, **84**, 162–169. 28

Wyatt, D.W., Li, H. & Tate, J.E. (2014). The impact of road grade on carbon dioxide (co 2) emission of a passenger vehicle in real-world driving. *Transportation Research Part D: Transport and Environment*, **32**, 160–170. 41, 56, 87, 125, 170, 174, 224, 251

Yang, L., Franco, V., Campestrini, A., German, J. & Mock, P. (2015a). Nox control technologies for euro 6 diesel passenger cars. *ICCT white paper (accessed on Apr., 2016) Search PubMed*. 46, 220

Yang, L., Franco, V., Mock, P., Kolke, R., Zhang, S., Wu, Y. & German, J. (2015b). Experimental assessment of no x emissions from 73 euro 6 diesel passenger cars. *Environmental science & technology*, **49**, 14409–14415. 46, 220

Yu, R., Wong, V. & Shahed, S. (1980). Sources of hydrocarbon emissions from direct injection diesel engines. Tech. rep., SAE Technical Paper. 60

Zallinger, M., Tate, J. & Hausberger, S. (2008). An instantaneous emission model for the passenger car fleet. In *16th International Transport and Air Pollution Congress*. 56

Zamboni, G., Malfettani, S., André, M., Carraro, C., Marelli, S. & Capobianco, M. (2013). Assessment of heavy-duty vehicle activities, fuel consumption and exhaust emissions in port areas. *Applied Energy*, **111**, 921–929. 56

Zhang, K., Batterman, S. & Dion, F. (2011). Vehicle emissions in congestion: Comparison of work zone, rush hour and free-flow conditions. *Atmospheric Environment*, **45**, 1929 – 1939. 21

Zhang, Y., Stedman, D.H., Bishop, G.A., Guenther, P.L., Beaton, S.P. & Peterson, J.E. (1993). On-road hydrocarbon remote sensing in the denver area. *Environmental science & technology*, **27**, 1885–1891. 61

Zhang, Y., Bishop, G.A. & Stedman, D.H. (1994). Automobile emissions are statistically gamma distributed. *Environmental science & technology*, **28**, 1370–1374. 6, 157

Zhang, Y., Stedman, D.H., Bishop, G.A., Guenther, P.L. & Beaton, S.P. (1995). Worldwide on-road vehicle exhaust emissions study by remote sensing. *Environmental science & technology*, **29**, 2286–2294. 65

ZHANG, Y., BISHOP, G.A., BEATON, S.P., GUENTHER, P.L. & MCVEY, L.F. (1996a). Enhancement of remote sensing for mobile source nitric oxide. *Journal of the Air & Waste Management Association*, **46**, 25–29. 61

ZHANG, Y., STEDMAN, D.H., BISHOP, G.A., BEATON, S.P. & GUENTHER, P.L. (1996b). On-road evaluation of inspection/maintenance effectiveness. *Environmental Science & Technology*, **30**, 1445–1450. 37, 61, 64