Study of biomass burning pollution over the Amazon using modelling and satellite data

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Abstract

Significant ozone levels are observed every year in the Amazon during the burning season, with potential risks for populations and ecosystems. The dynamics that govern the distribution of biomass burning pollution across the region and hence, the impact on surface ozone are still unknown. Fire activity is predicted to increase, due to its strong dependence on global warming and droughts. Thus, understanding the vertical distribution of biomass burning emissions in the Amazon is crucial to determine and quantify the impacts. For that, this work used satellite observations, aircraft and ground-based measurements and ozonesondes, combined with an Earth system model.

The first part of this work characterised the vertical distribution of Amazonian smoke plumes from satellite observations and analysed major factors of variability. The statistical analysis of smoke plume characteristics combined with an extensive dataset on the main drivers in smoke plume dynamics revealed that most smoke concentrates below 2.5 km and plume heights depend largely on biome type, fire properties, and atmospheric and drought conditions. Specifically, droughts enhanced fire activity, favoured lower smoke plume heights and larger emissions, which may result in poor regional air quality with important implications in the future, when more severe and extended droughts are expected.

To improve the vertical distribution of biomass burning pollution across the Amazon in Earth system models, an injection height scheme derived using observations of smoke plumes in the Amazon was applied. The simulation showed better performance at representing ozone compared to observations, particularly close to the fires. Furthermore, results evidenced a significant impact of biomass burning emissions on ozone levels, and a considerable decline in air quality across populated and vegetated areas. These outcomes highlighted the necessity of including improved representation of the vertical distribution of biomass burning emissions in future air quality studies and provided insights of the magnitude of biomass burning impact on air quality, enhancing scientific understanding of the significance of biomass burning in the Amazon.

Dedication

To the memory of Armando and my Dad.

Declaration

I, Laura Gonzalez Alonso, hereby declare that I am the sole author of this thesis and that, except where specific reference is made to the work of others, the contents of this research is the result of my own work, unless otherwise acknowledged in the text. I confirm that this dissertation is original and has not been submitted for any other degree, diploma or other qualifications to this university or any other institution.

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Chapter 1

Introduction

1.1 Background

Biomass burning is the combustion of living and dead vegetation, including natural and anthropogenic burning. Every year, vegetation fires burn around 3 million km^2 of land globally (Giglio et al., 2010), which constitutes a significant primary source of gases and particles (Crutzen and Andreae, 1990, Andreae and Merlet, 2001, Ito and Penner, 2004, van der Werf et al., 2006, Wiedinmyer et al., 2010), equivalent to about 20% of global emissions from fossil fuels (Denman et al., 2007), and contributes to the formation of secondary pollutants (Val Martin et al., 2006, Alvarado et al., 2010, Akagi et al., 2011, Jaffe and Wigder, 2012). Gases released by fires include greenhouse gases i.e., carbon dioxide (CO₂), methane (CH₄) and nitrous oxide (N₂O), reactive trace gases i.e., sulfur oxides (SO_x) and ammonia (NH₃), some of which are precursors of tropospheric ozone (O₃), i.e., carbon monoxide (CO), volatile organic compounds (VOC), nitrogen oxides (NO_x). Fine and coarse particulate matter (PM) are also largely produced by fires (Goode et al., 2000, Andreae and Merlet, 2001), including black and organic carbon.

Biomass burning emissions significantly influence the chemical composition of the atmosphere (e.g., Yurganov et al., 2004, Lapina et al., 2006, Simpson et al., 2006), with the potential to degrade air quality, being detrimental to human health and ecosystems, as well as to reduce visibility (e.g., Lippmann, 1993, Fiscus et al., 2005, Felzer et al., 2007, Jaffe et al., 2008, Ainsworth et al., 2012, Marais et al., 2014, Reddington et al., 2015). For instance, in a recent comprehensive 20-year modelling study, Jacobson (2014) suggested that biomass burning may be responsible for around 250,000 premature deaths per year. In addition, fire emissions alter weather and climate from regional to global scales (e.g., Ramanathan et al., 2001, Yurganov et al., 2004, Langmann et al., 2009) directly, via emitting greenhouse gases and aerosols, and increasing the amount of solar radiation absorved or reflected to space (Ramanathan and Carmichael, 2008), and indirectly, via secondary effects on atmospheric chemistry (e.g., ozone and secondary organic aerosols formation) or changes in cloud microphysics, precipitation regimes and albedo, from aerosol emissions (e.g., Twomey, 1977, Albrecht, 1989, Sitch et al., 2007). Thus, emissions from biomass burning have been suggested to cause a 20-year global warming of 0.4 K (Jacobson, 2014).

Biomass burning contributes substantially to global CO, NO_x and O_3 budgets (32%, 21% and 3.5%, respectively) (Andreae, 1991, Jaffe and Wigder, 2012). CO and NO_x play an important role in atmospheric chemistry. CO acts as the dominant sink for the hydroxyl radical (OH), the main tropospheric oxidant, with 90-95% of CO and about 75% of OH removal (Novelli et al., 1998). In addition, CO oxidation provides a source for O_3 formation in the presence of NO_x (e.g., Levy, 1971, Crutzen, 1973, Logan et al., 1981). NO_x promote substantial changes in the chemical production and loss rates of O_3 and CH_4 (Andreae and Merlet, 2001). Therefore, emissions of CO and NO_x have the potential to influence air quality and climate by altering CH_4 and other radiatively important gases that are removed by OH, and by affecting tropospheric O_3 itself (e.g., Mickley et al., 1999).

Tropospheric O_3 is an important oxidant (Seinfeld and Pandis, 1998) and source of OH. In addition, O_3 is the third most important greenhouse gas (IPCC, 2007) and a major air pollutant harmful to human health and plants. Inhaled O_3 can cause decreases in lung function, aggravation of asthma, throat irritation and cough, chest pain and shortness of breath, inflammation of lung tissue, higher susceptibility to respiratory infection and premature mortality (e.g., Bell et al., 2004, 2006, Kheirbek et al., 2013, Liu et al., 2018). Exposure to ozone can also cause a range of effects on vegetation, including visible leaf injury (e.g., Fumagalli et al., 2001), growth and yield reductions, and altered sensitivity to abiotic and biotic stresses, such as droughts and funghi, respectively (e.g., Fuehrer and Achermann, 1994, Ashmore and Marshall, 1998, Benton et al., 2000). For instance, increases in tropospheric O_3 due to fire emissions have been found to reduce global forest net primary production by 0.7% per year, considerably larger than reductions from droughts (0.1% per year) (Yue and Unger, 2018). Exposure to O_3 has also been found to produce crops yield losses (e.g., Avnery et al., 2011), resulting in substantial economic costs and posing a risk to global food security (Van Dingenen et al., 2009b, Avnery et al., 2011, Ghude et al., 2014). O_3 forms from the photochemical reaction through the oxidation of CO, CH_4 and VOCs, controlled and catalyzed by NO_x (NO_x denotes the sum of NO and NO₂) (Jonson et al., 2006). In urban areas, NO_x is mainly emitted by the combustion of fossil fuels, whereas in rural areas NO_x is produced by biomass burning, peroxyacetyl nitrate (PAN) decomposition and soils (Jaeglé et al., 2005). Most of the direct emission of NO_x is in the form of NO, which is rapidly transformed into NO₂ (~ 5 min) (Seinfeld and Pandis, 1998). VOCs are mainly produced by plants and to a lesser extent, by a range of industrial activities, road traffic and fires (Lerdau et al., 1997).

Ozone production starts with the photolysis of NO_2 (Reaction 1.1) and subsequent reaction of the oxygen atom with molecular oxygen (Reaction 1.2).

$$NO_2 + hv \longrightarrow NO + O$$
 (1.1)

$$O + O_2 + M \longrightarrow O_3 + M$$
 (1.2)

Once formed, ozone reacts with NO to form NO_2 .

$$O_3 + NO \longrightarrow NO_2 + O_2$$
 (1.3)

Reactions 1.1, 1.2 and 1.3 constitute a net zero ozone production cycle. However, in the presence of ozone precursors such as CO, VOC and hydrocarbons, net ozone production occurs. The chemistry leading to O_3 production starts with the oxidation of CO by the OH radical, forming the peroxy radical (HO₂) (Reaction 1.4). The formed HO₂ converts NO to NO₂ (Reaction 1.5), and photolysis of NO₂ forms O₃ (Reactions 1.1 and 1.2).

$$CO + OH \xrightarrow{O_2} HO_2 + CO_2$$
 (1.4)

$$HO_2 + NO \longrightarrow NO_2 + OH$$
 (1.5)

In the case of VOC reactions are

$$\operatorname{VOC} + \operatorname{OH} \xrightarrow{\operatorname{O}_2} \operatorname{RO}_2 + \operatorname{H}_2\operatorname{O} \tag{1.6}$$

$$\operatorname{RO}_2 + \operatorname{NO} \xrightarrow{\operatorname{O}_2} \operatorname{Secondary} \operatorname{VOC} + \operatorname{HO}_2 + \operatorname{NO}_2$$
 (1.7)

and subsequent Reactions 1.5, 1.1 and 1.2 to form O_3 .

The ratio of VOCs to NO_x is particularly important in ozone formation. In VOCs-limited environments (e.g. urban areas or polluted remote areas), increases in VOCs leads to higher O_3 production, but lower O_3 production if NO_x mixing ratio increases above 300 ppt (NO_x -titration) (Sillman, 1999). In NO_x -limited environments (e.g. the Amazon), O_3 production increases with increasing NO_x and shows relatively little change in response to increased VOC.

In biomass burning plumes O_3 formation is complex, non-linear and highly variable, depending on many factors, such as photochemical conditions, ageing and dilution/mixing (Jaffe and Wigder, 2012). Some studies have shown rapid production

of O_3 in plumes (Baylon et al., 2015), whereas others have shown no enhancement or even depletion (Alvarado et al., 2010, Akagi et al., 2011, Baylon et al., 2015, Verma et al., 2009). One reason for the difference in O_3 formation rates in plumes is the variability in NO_x/CO ratios among fires. This ratio is lower (higher) for fires with dominant smouldering (flaming) combustion (Lobert and Warnatz, 1993, Yokelson et al., 1997, Goode et al., 2000) however, O_3 production in the plume can be enhanced downwind in the presence of additional sources of NO_x , as the plume encounters polluted air from urban areas (Singh et al., 2010). Another reason for differences in O_3 production in plumes is the presence of aerosols (Baylon et al., 2018), which can reduce O_3 photochemical reactions due to aerosol absorption and scattering of solar radiation (Xing et al., 2017).

The atmospheric impacts of biomass burning, e.g., O_3 production, depend on many factors: the amount of emissions released for each species, meteorological conditions, topography, factors related to fire behaviour (fire intensity, fuel availability, fuel characteristics, i.e., type, loading, moisture) and injection height, i.e., the altitude at which fire emissions are released. A key factor in biomass burning emissions is the combustion process, which is directly related to the type and amount of species emitted and their vertical distribution. Combustion can be divided into several stages: distillation or drying, pyrolysis, flaming combustion and smouldering combustion (Benkoussas et al., 2007), which in vegetation fires usually occur simultaneously and in the immediate surroundings. In terms of fire emissions, combustion is mainly divided into flaming and smouldering combustion and their ratios in a fire vary over time. Typically, flaming dominates in the earlier stage of a fire, whereas smoldering occurs in a later stage (Andreae and Merlet, 2001). Flaming combustion is characterised by intense flames, higher rates of spread, and high temperatures $(\sim 1500^{\circ}\text{C})$ (Rein, 2016), which produce gas-phase emissions dominated by highly oxidised compounds (i.e., CO_2 , NO_x) (Lobert et al., 1991, Yokelson et al., 1997, Radke et al., 1991, Reid et al., 2005, Chen, 2007). Temperate forest, tropical savannas and grassland fires are typically dominated by flaming combustion. On the other hand, smouldering combustion is the slow, persistent, low-heat (450–700°C) flameless burning (Rein, 2016), which releases incomplete combustion products (i.e., CO). Areas with soil rich in organic matter and high moisture content, i.e. boreal forest, tropical forest and peatland, are mainly dominated by smouldering fires.

The injection height is another important factor in the atmospheric impacts of biomass burning. It is directly linked to the combustion stage and determines the lifetime and behaviour of the emitted species (Freitas et al., 2006, Paugam et al., 2016). Low-intensity smouldering fires tend to produce weaker buoyant smoke plumes than intense flaming fires (Val Martin et al., 2010, Amiridis et al., 2010). When a fire is in its flaming combustion stage, the intense heat released from the burning creates fire-induced convection above the fire, and a buoyant smoke plume originates, which interacts with the ambient atmosphere and transports fire emissions vertically. Most fire plumes concentrate below the planetary boundary layer (PBL), where emissions are well-mixed (Trentmann et al., 2002) and their impacts extend on a local to regional scale. However, a significant fraction of smoke can reach the free troposphere (FT) (e.g., Kahn et al., 2008, Val Martin et al., 2010), extending the lifetime of the emitted species and the spatial scale of their impact, due to faster downwind transport (Fromm et al., 2004). Injection heights in smoke plumes are highly variable. Atmospheric conditions and the energy released by the fire are the main drivers of the variability associated with smoke plume heights (Kahn et al., 2007, Paugam et al., 2016). For example, the thermal stratification of the atmosphere can promote or suppress the plume rise. That is, if the atmospheric temperature at a certain level is lower than the plume's, the plume tends to ascend. Furthermore, in the case of energetic fires, and in the presence of water vapour condensation and latent heat release, the vertical transport within the plume can be invigorated, and occasionally form pyro-cumulus clouds that inject large amounts of biomass burning emissions into the FT, even reaching the lower stratosphere (Fromm et al., 2010).

Plume-rise models (PRM) and semi-empirical parametrisations use atmospheric

profiles of meteorological variables combined with satellite observations related to the fire, i.e., fire size, fire radiative power (FRP) to predict the evolution of a plume (Paugam et al., 2016, Rémy et al., 2017). They are usually included in chemical transport models (CTM) and provide results on the injection heights, but simulations are computationally expensive and poorly validated, particularly for PRM (Val Martin et al., 2012). Satellite observations, ground-based and aircraft measurements provide more accurate data on the vertical distribution of smoke. This information is commonly used to evaluate results from PRM, as well as to constrain the vertical distribution of biomass burning emissions in CTM. Across the globe, many studies have sought to characterise smoke plume heights. Table 1.1 presents a summary of the most relevant to this study.

Many pollutants released or produced in fires are regulated due to their negative effects on human health and ecosystems, including O_3 (Felzer et al., 2007, Wegesser et al., 2009, Haikerwal et al., 2016, Crippa et al., 2016, Schweizer and Cisneros, 2017). O_3 standards and regulations vary substantially depending on the country and region, and provide guidelines and limit values to safeguard human health and ecosystems based on some metrics. Some of the most commonly used metrics for O_3 air quality regulations are summarised in Table 1.2. O_3 exposure risks for human health are typically assessed with MDA8 as the basic metric. MDA8 can be applied in combination with a number of exceedances that are allowed before violation of O_3 standards occurs (Fleming et al., 2018). For instance, the European Commission (under Directive 2008/50/EU) has a target value for MDA8 ozone concentrations of 60 ppb not to be exceeded on more than 25 days per calendar year. A summary of the main O_3 exposure standards for human health, including those relevant to this study, are presented in Table 1.3. Previous studies that used some of these standards to assess air quality, suggested that intense biomass burning periods can significantly increase the frequency of O_3 standards exceedances (Jaffe et al., 2008, Pfister et al., 2008, Chalbot et al., 2013, Rubio et al., 2015, Brey and Fischer, 2016). However, most of these studies have focused on fire-induced exceedances across the

$Parameter^{a}$	Smoke height ^{b}	Temporal	Spatial	Instrument/	Reference
		coverage	$coverage^{c}$	$\mathrm{Product}^d$	
Att. bck.	<pbl< td=""><td>Jul-Aug 2006</td><td>GFR</td><td>CALIOP L1</td><td>Labonne et al. (2007)</td></pbl<>	Jul-Aug 2006	GFR	CALIOP L1	Labonne et al. (2007)
Plume	$0.7{-}5.2~\mathrm{km}$	Jun-Sep 2004	North America	MISR/MODIS	Mazzoni et al. (2007)
Plume	$0.18{-}4.5~\mathrm{km}$	summer 2004	Alaska-Yukon	MISR/MINX	Kahn et al. (2008)
CO	10-20% FT	Jun–Oct 2006	GFR	TES /MLS	Gonzi and Palmer (2010)
AI/Att. bck.	AI>9; >5 km	2006 - 2009	Globe	OMI/CALIOP L2	Guan et al. (2010)
Plume	$0.7{-1}$ km	2002/2004-2007	North America	MISR/MINX	Val Martin et al. (2010)
Att. bck.	1.65.9 km	2006-2008	SW Russia/E Europe	CALIOP L1	Amiridis et al. (2010)
Plume	$26\%>\mathrm{PBL}$	$\mathrm{Dec}2000/\mathrm{Nov}2002$	Australia	MISR/MINX	Mims et al. (2010)
Plume	$0.7 \mathrm{km}$	2001 – 2009	Borneo/Sumatra	MISR/MINX	Tosca et al. (2011)
Ext. coef.	0.5-4 km	2006 – 2009	Borneo	CALIOP L1	Tosca et al. (2011)
Att. bck./Ext. coef.	$<\!2/3$ –5 km	Jul–Nov 2008	Manaus, BR	Raman lidar	Baars et al. (2012)
Plume	45%>1 km	2001 - 2010	SE Asia	MISR/MINX	Jian and Fu (2014)
AOD	1.6-4 km	2007 - 2012	GFR	CALIOP L2	Huang et al. (2015)
Extinction	$2-5 \mathrm{~km}$	2007 – 2012	South America	CALIOP L2	Bourgeois et al. (2015)
Att. bck.	$0.8{-}5.3~\mathrm{km}$	2005 – 2012	Asia	CALIOP LI	Vadrevu et al. (2015)
Ext. coef.	$1{-}1.5/4{-}6~{ m km}$	16-29 Sep 2012	6 flighttracks, BR	ALS-450 lidar	Marenco et al. (2016)
Plume/AOD	$<2~{ m km}$	2008 - 2010	Globe	MISR/MINX	Val Martin et al. (2018b)

Table 1.1: Summary of the most relevant observational studies on smoke heights across the globe.

ate between smoke layers, when observed more than one. GFR: global fire regions; BR: Brazil. ^bSmoke height is given as a range of heights where smoke concentrates, as a percentage above or below the PBL or the FT. / is used to separ-

^dCALIOP: Cloud-Aerosol Lidar with Orthogonal Polarization; L1: level 1; L2: level 2; MISR: The Multi-angle Imaging SpectroRadiometer; MODIS: Moderate Resolution Imaging Spectroradiometer; MINX: MISR INteractive eXplorer; TES: The Tropospheric Emission Spectrometer; MLS: the Microwave Limb Sounder; OMI: Ozone Monitoring Instrument.

northern hemisphere (NH). In addition to human health standards, O_3 metrics for vegetation i.e., AOT40, W126 and M12, are used to determine levels above which adverse effects on sensitive vegetation may occur. Table 1.4 summarises some of the most relevant. Studies on the global impact on crops yield of current and future exposure to elevated concentrations of ozone suggested substantial yield reductions, depending on crop and metric (3-16%), enhanced under future scenarios (by >10%) (Van Dingenen et al., 2009b, Avnery et al., 2011, Tai et al., 2014). As in the case of ozone impacts on human health, most of these studies are based on dose-response functions for agricultural and horticultural crops in the NH.

Table 1.2: Summary of metrics relevant to ozone standards for air quality. *n* is the number of hours in the growing season, $[O_3]$ is the hourly ozone concentration from 08:00–19:59 hours and *i* is the hour index.

Metric	Definition	Unit	Application
MDA8	maximum daily 8-h mean	ppb	human health
AOT40	$\sum_{i=1}^{n} \left[[O_3] - 0.04 \right]_i$	ppm h	vegetation
	for $[O_3] \ge 0.04 \text{ ppm h}$		
W126	$\sum_{i=1}^{n} \left[\frac{[O_3]}{1+4403 \exp(-0.126 \times [O_3])} \right]_i$	ppm h	vegetation
	for $[O_3] \ge 0$ ppm h		
M12	$\frac{1}{n}\sum_{i=1}^{n}[O_3]_i$	ppb	vegetation

Table 1.3: Summary of relevant ozone standards for human health. (Adapted from Fleming et al. (2018)).

$\operatorname{Region}^{a}$	$Metric^{b}$	Value [ppb]	Reference
WHO	MDA8	55	WHO (2008)
EU	MDA8	60^{c}	CLRTAP (2017)
USA	4MDA8	70	EPA (2016)
AMAZON	MDA8	$40 - 80^{d}$	National environmental agencies ^{e}

^a WHO: world Health Organization; AMAZON: Bolivia, Brazil, Colombia, Chile, Ecuador, French Guiana, Peru, Venezuela.

^b 4MDA8: annual 4th highest MDA8.

^c not to be exceeded on more than 25 days per calendar year.

^d MDA8 standards range across the Amazon region.

e http://www.cleanairinstitute.org; https://www.minambiente.gov. co/; http://www.leychile.cl; http://www.mma.gov.br

Intense biomass burning in the Amazon contributes to the global fire emissions by approximately 15% (Van der Werf et al., 2010, Mishra et al., 2015), so the Amazon

Metric	Period	Standard	Vegetation	Effect/	Reference
			type	Reduction	
	3-months	3000 ppb h	Agricultural	Grain yield $(5\%^a)$	
	3-months	8000 ppb h	Horticultural	Fruit yield $(5\%^b)$	-
AOT40	6-months	5000 ppb h	Forest	Biomass $(5\%^c)$	CLRTAP
	3-months	3000 ppb h	Annuals	Biomass (10%)	(2017)
	6-months	5000 ppb h	Perennials	Biomass (10%)	-
		15000 ppb h	Crops	Prevent loss $>5\%$	
W126	Highest	17000 ppb h		Improve protection	US Federal
	consecutive	10000 ppb h	Tree Plants	Reduce foliar injury	Register
	3-months	7000 ppb h	Ecosystems	Limit loss $< 2\%$	(2015)

Table 1.4: Summary of ozone main standards for vegetation. (Adapted from Mills et al. (2018)).

^a Based on wheat.

^b Based on tomato.

^c Based on beech and birch.

is one of the most important biomass burning regions in the world. With one of the largest global deforestation rates (Artaxo et al., 2002, Malhi et al., 2008), every year, thousands of fires burn in the Amazon basin. Most fires are of anthropogenic origin, i.e., for preparation of agricultural or pastoral lands, and burn during the burning season, from July to November, across the arc of deforestation (Andreae et al., 2012) (see Figure 1.1), with dominant burning of savanna and tropical forest (94%) of the fires) (Gonzalez-Alonso et al., 2019). Furthermore, the Amazon basin covers an area of about 35.5% of South America and comprises the countries of Bolivia, Brazil, Colombia, Ecuador, Guyana, Peru, Suriname and Venezuela, with a population of 25 million people (Davidson et al., 2012), including indigenous communities, unique biodiversity and a rich agriculture-based economy (i.e., cocoa, coffee, quinoa). Thus, a large portion of this population suffers regularly from high level of pollutants from biomass burning emissions (Brito et al., 2014). At the same time, the Amazon basin contains the world's largest rainforest (Laurance et al., 2001, Aragao et al., 2014), which is a key component of the Earth System. It provides about a fifth of all of the freshwater inputs to the global oceans (Marengo and Espinoza, 2016, Nobre et al., 2016), which makes it the single, largest source of fresh water on the Earth. The Amazon rainforest stores approximately 120 billion tonnes of carbon (Malhi et al., 2006, Saatchi et al., 2011), equivalent to approximately 9-14 decades of current global anthropogenic carbon emissions (Canadell et al., 2007), and absorbs about 1 billion tonnes of carbon per year (more than 10% of annual anthropogenic CO_2 emissions) (Marengo et al., 2018). In addition, moisture exchanges in the Amazon forest play a crucial role in the climate system, contributing to atmospheric circulation and to the water, energy and carbon cycles (Zemp et al., 2014, Spracklen and Garcia-Carreras, 2015, Nobre et al., 2016). However, climate variability and anthropogenic activities, i.e., deforestation fires, have become important agents of disturbance in the Amazon basin (Davidson et al., 2012).



Figure 1.1: Satellite image collected by the Moderate Resolution Imaging Spectroradiometer (MODIS) aboard the Terra satellite on August 18, 2015. Actively burning areas, detected by MODIS, are outlined in red and the arc of deforestation shaded in red; Image adapted from https://www.nasa.gov/image-feature/goddard/ el-ninos-effects-bring-more-wildfires-to-brazil.

Previous studies have sought to understand the impact of biomass burning emissions in the Amazon from local to hemispheric scales (Andreae et al., 1988, Kirchhoff

et al., 1989, Zhang et al., 2008, Ignotti et al., 2010, de Andrade Filho et al., 2013, Kolusu et al., 2015, de Oliveira Alves et al., 2015, Reddington et al., 2015, Archer-Nicholls et al., 2016, Martin et al., 2016, Giangrande et al., 2017). For instance, a few studies on smoke height across the Amazon have determined that smoke tends to concentrate under 2.5 km although they also found the presence of a persistent haze layer at around 4–6 km (Table 1.1). These studies are based on limited observations for short periods of time or specific locations, that may be influenced by specific weather conditions. Because of the lack of resources and complexities of such a complex, vast and undeveloped area, in-situ sampling in the region is scarce. Therefore, studies on biomass burning across the region have typically used satellite observations (i.e., MOPITT, MAPS and TOMS), ozonesondes and ground-based observations, combined with global and regional CTMs supplied with meteorological data and fire emission estimations. In addition, aircraft campaigns across the region have been designed to overcome the scarcity of observations, by providing with high temporal and spatial resolution data on biomass burning pollution, but limited to flight tracks. These include ABLE 2A (Harriss et al., 1988), CITE 3 (Hoell Jr et al., 1993), TRACE A (Fishman et al., 1996b), BARCA (Andreae et al., 2012), and more recently, SAMBBA (Allan et al., 2014) and GoAmazon (Martin et al., 2016). Overall, these observational and modelling studies have revealed that emissions from biomass burning in the Amazon are a large contributor to CO and O_3 budgets and their interannual variability in the southern hemisphere (SH), as well as they have shown high mixing ratios of both gases in the mid-upper troposphere over the region (e.g., Reichle et al., 1986, Andreae et al., 1988, Kirchhoff and Rasmussen, 1990, Watson et al., 1990, Fishman et al., 1996a, Galanter et al., 2000, Thompson et al., 2001, Edwards et al., 2006, Deeter et al., 2018). Substantially high surface CO and O_3 mixing ratios of 400 ppb (Andreae et al., 2012) and 40–60 ppb (Bela et al., 2015), respectively, have also been reported during the burning season, even reaching maximum daily surface O₃ mixing ratios as large as 100 ppb (Artaxo et al., 2002, Kirkman et al., 2002). The ozone levels found are well above the critical level known to be hazardous to human health and plants (40 ppb) (Ainsworth et al., 2012). Furthermore, Pacifico et al. (2015) assessed the impact of fire-induced ozone exposure on the Amazonian tropical forest productivity and suggested enhancements of 15 ppb in O_3 mixing ratios, due to biomass burning, which resulted in mean reductions in forest productivity of 15%. Nevertheless, modelling studies across the Amazon have reported some systematic quantitative differences compared to observations, which seemed to be related to poor representation of biomass burning emissions and smoke injection heights, as well as convective and long-range transport in the models (Andreae et al., 2012, Bela et al., 2015).

Despite the large influence of biomass burning from the Amazon on the atmosphere budget, and the critical levels of ozone found each year during the burning season, no studies have yet comprehensively investigated the smoke plume dynamics governing the region, or assessed biomass burning impacts on surface ozone levels, with implications for human health and crops productivity. Future projections suggest an increase in fire activity over the Amazon region (Cochrane and Barber, 2009), exacerbated by more frequent droughts, as a consequence of climate change and human activities (Bowman et al., 2009). Under this scenario, increases of fire emissions are expected, which may lead to large, more frequent and extended episodes of ozone pollution, compromising larger population's health and food security. To fully understand the factors that drive smoke plume dynamics and the transport and distribution of pollution produced in a fire is crucial to accurately predict and help mitigate impacts on air quality and climate, from local to global scales, as well as minimise the risks to human population and ecosystems.

1.2 Motivation, research objectives and approach

By 2015, an estimated area of 66% of the total Brazilian Amazonia had been deforested (INPE, 2016). Extensive deforestation leads to changes in Amazon forest dynamics with the potential to affect the concentration of atmospheric CO₂ and modify precipitation, among other parameters (i.e., albedo, temperature, surface roughness, stomatal resistance, soil moisture). All these changes have significant consequences on global climate, i.e., air cooling and changes in large-scale circulation (Nobre et al., 1991, Marengo and Nobre, 2001, Werth and Avissar, 2002). Furthermore, deforestation fires in combination with global warming and more frequent and severe droughts may increase biomass burning emissions and the Amazon forest may become in the near future, a source of carbon rather than a sink (Davidson et al., 2012).

In view of the importance of the Amazon as a global stabiliser and the large contribution of local biomass burning emissions to the global and regional atmospheric budget, it is crucial to have a better understanding of the drivers that control the transport and distribution of biomass burning pollution over the Amazon, its contribution to the atmospheric composition and its global and regional impacts. This project seeks to characterise smoke plume dynamics across the region, which will help represent the best modelling approach to study biomass burning over the Amazon, assess the contribution of biomass burning to the ground ozone levels and associated impacts on air quality. For this purpose, satellite observations, ozonesondes, and aircraft and ground-based measurements combined with a global Earth System Model (ESM) are employed. Specifically, this study seeks to answer the following scientific questions:

What is the vertical distribution of biomass burning emissions over the Amazon? Determining the height at which fires inject pollutants in the atmosphere will allow understanding of how and in which degree biomass burning in the Amazon impacts the atmospheric composition, air quality and climate, from regional to global scales. Despite the importance of fire emissions from the Amazon in the global atmospheric budget, little is known about the processes that control fire pollution and plume dynamics over this region, mostly due to the lack of smoke plume height observations. This study proposes the use of a combination of satellite data during the burning seasons of 2005-2012 to develop a climatology of smoke plume heights over the Amazon. The information obtained from this analysis will help better

represent the vertical distribution of Amazonian fire emissions in ESMs.

Which are the main factors that control the vertical distribution of biomass burning emissions over the Amazon? Biomass burning in the Amazon is influenced by complex interactions among meteorology, climate, topography and human activities. Identifying main drivers of variability in fire plume dynamics across the Amazon is key to define future trends and make decisions to responsibly manage air quality and climate. On that respect, this project will evaluate the main aspects that affect smoke plume dynamics. This includes an extensive evaluation of fire properties, plume characteristics, weather and climatic conditions from 2005 to 2012.

What is the influence of biomass burning on surface O_3 levels and its impact on air quality over the Amazon? Exploring the contribution of Amazonian biomass burning emissions on surface ozone levels and its potential toxicity and phytotoxicity is crucial to understanding the regional and large-scale implications on air quality. For this, results from scientific questions 1 and 2 will provide information to better represent the vertical distribution of biomass burning emissions in an ESM, and assess the potential contribution of biomass burning to surface ozone levels and the impacts on air quality. This work will implement a novel fire injection height parametrisation, based on satellite observations, into an ESM and evaluate results with a combination of satellite observations, ozonesondes, aircraft and ground-based measurements. Finally, results from the modelling experiments will help widen the understanding of the impacts of fire-induced ozone on human health and vegetation across the region.

1.3 Dissertation overview

The following chapters include data, methods, analyses, results and conclusions that address the research objectives of this study. Chapter 2 presents an overview of the main features, settings, performance and limitations of the software used to develop a climatology of smoke plumes across the Amazon for 2005-2012. This chapter addresses the first research objective. Chapter 3 presents an analysis of the climatology of smoke plume heights derived from satellite observations and assesses the main drivers of variability on smoke plume heights across the region, which directly addresses first and second research objectives. This chapter is included as a manuscript that was published on February 8th, 2019 in the Atmospheric Chemistry and Physics journal (ACP). Chapter 4 presents a modelling analysis of the impact of the vertical distribution of biomass burning on ozone and its precursors and assess the influence of Amazonian biomass burning on surface ozone and air quality over the Amazon region. This chapter is inserted as a manuscript to be submitted to ACP. Chapter 5 provides conclusions from all the analyses conducted in this work and recommendations for future research. Appendix A includes supplementary information for chapter 3. Appendix B includes supplementary information for chapter 4. Appendix C presents a summary of the chapters with contributions.

Chapter 2

MISR and MINX: Developing a biomass burning smoke plume climatology across the Amazon

2.1 Introduction

Remote sensing techniques allow observing the spatial and temporal distribution of aerosols in the atmosphere, which is crucial to study their impacts on climate and air quality. Passive remote sensing techniques detect the natural radiation reflected or emitted by features under cloud-free conditions. They provide high spatial and temporal coverage, but limited accuracy on the vertical aerosols distribution. These passive techniques include Radiometry, Imaging Radiometry, Spectrometry and Spectroradiometry. The latter is used by the Multi-angle Imaging SpectroRadiometer (MISR) combined with multi-image matching stereoscopic techniques, based on the principle of parallax (Diner et al., 1998). An important advantage of this technique is that it relies uniquely on geometry and no calibration is needed, but its major limitation is its low sensitivity to thin aerosol features without a well-defined contour that is not clearly discernible from the background. On the other hand, active remote sensing techniques send a pulse of energy and receive the radiation reflected. These techniques include Radar, Scatterometry, Laser altimetry and LIDAR, such as the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP), on board the CALIPSO satellite. CALIOP provides high accurate aerosol scattering profiles (Winker et al., 2009) but extremely low spatial coverage due to its narrow path (\sim 60 m).

MISR and CALIOP have been widely used in the study of the vertical distribution of aerosol plumes and clouds in the atmosphere over several regions (Val Martin et al., 2010, Amiridis et al., 2010, Jian and Fu, 2014, Huang et al., 2015). For instance, over North America, Val Martin et al. (2010) developed an extensive 5-year climatology of smoke plume heights based on height-retrievals derived using MISR imagery. Similarly, Jian and Fu (2014) and Tosca et al. (2011) characterised smoke plume heights during the burning seasons of 2001-2009/2010, over tropical regions in Asia and Mims et al. (2010), over grassland fires in Australia. Using observations made by CALIOP, Huang et al. (2015) examined the most probable height of dust and smoke layers over six fire impacted regions and Amiridis et al. (2010)investigated aerosols vertical distribution and smoke top heights from agricultural burning in Europe. All these studies showed the large variability in smoke plumes across biome, season and region, as well as demonstrated that although most smoke concentrates in the boundary layer, where it is well-mixed, a variable but significant percentage of generally, low-density smoke reaches the free troposphere, as a result of favourable fire and local weather conditions, and can be transported long-range distances. MISR and CALIOP performance and sensitivity are disparate. Specifically MISR provides near-source constraints on the vertical distribution of smoke and allows to study smoke plume dynamics on a plume-by-plume case.

The Amazon region is a major fire region, which contributes largely to the global fire emissions (Van der Werf et al., 2010). However, despite its important role in the distribution and transport of global biomass burning products, no study has yet developed a climatology of smoke plume heights over the region. The present study
aims at improving the vertical distribution of biomass burning emissions represented in Earth system models (ESM) over the Amazon. For that, MISR capabilities are exploited to develop a large dataset of smoke plume heights during the burning seasons (July to November) from 2005–2012. This is the first time that such a comprehensive study of the vertical distribution of biomass burning emissions has ever been done over the Amazon. The smoke plume database developed over the Amazon presented in Chapter 3 (Gonzalez-Alonso et al., 2019) was created with the MINX interactive tool (Nelson et al., 2008b, 2013), using the MISR imagery and MODIS thermal anomalies (Diner et al., 1998, Giglio et al., 2003). Because the use of MINX requires some understanding of the software and algorithms used, this chapter describes the principal features of the MISR instrument and performance of MINX, with focus on the Amazon smoke plume climatology.

2.2 MISR Instrument and products

The Multi-angle Imaging SpectroRadiometer (MISR) is a spaceborne instrument that measures atmospheric and surface properties, designed to study cloud, aerosols and the Earth surface. MISR flies on board the Terra satellite (launched in December 1999) and since February 2000 has been acquiring images of the Earth at nine different fixed angles (from -70°to 70°)(Diner et al., 1998). This multi-angle imagery provides stereoscopic retrievals of aerosol plumes and clouds heights at 275 m to 1.1 km of resolution. MISR is integrated in the NASA Earth Observing System (EOS) fleet, with a near-polar orbit at an altitude of 705 km and about 380 km of swath common to all cameras. The descending node crosses the Equator at around 10:30 a.m. local time and provides global coverage every 9 days at the equator and every two days near the poles.

The nine push-broom cameras are designated as An, for the nadir camera and A, B, C and D followed by "a" or "f", depending of their viewing, being "a" for aft-viewing and "f" for forward-viewing (i.e., Aa, Ba, Ca, Da and Af, Bf, Cf Df;

Figure 2.1). The camera viewing angles differ in approximately 7 min from each other and $70.5^{\circ}(D)$, $60.0^{\circ}(C)$, $45.6^{\circ}(B)$, and $26.1^{\circ}(A)$ from the nadir. Images are acquired in four spectral channels for each camera at blue, green, red and near-infrared wavelengths (446.4 ± 41.9 ; 557.5 ± 28.6 ; 671.7 ± 21.9 and 866.4 ± 39.7 nm, respectively). More information about the MISR instrument can be found in Diner et al. (1998) and at https://misr.jpl.nasa.gov/Mission/misrInstrument/.



Figure 2.1: TERRA satellite with MISR aboard, and the four-spectral-band multi-angle nine cameras; https://github.com/nasa/MINX/blob/master/webdoc/MINX_Doc1.pdf.

MISR data are freely downloadable from the Earth Data website¹, following registration and logging in. Data include three level products and ancillary data. The Levels 1 and 2 products are in swaths of 180 blocks of 140.8 km along track for each MISR orbit. Level 1 products have been processed and calibrated radio-metrically and geometrically to remove many of the instrument effects. Level 2

¹https://urs.earthdata.nasa.gov; last access 14/02/2019

products are geophysical measurements derived from the instrument data. They include Level 2 Top-of-Atmosphere/Cloud product, with cloud heights and winds, cloud texture, top-of-atmosphere albedos and other related parameters, and Level 2 Aerosol/Surface product, with tropospheric aerosol optical depth, aerosol composition and size, among other parameters. Level 3 product consists of monthly, seasonally, and annually averaged maps for various parameters from Level 2. The NASA Langley Atmospheric Science Data Center² distributes the MISR products in hierarchical data format (HDF). Detailed description of the algorithms used for each data product and specifications of the HDF files can be found in the Atmospheric Science Data Center (ASDC) website³.

The stereo-matching algorithms in the operational MISR Level 2 cloud product use MISR multi-angle ellipsoid-referenced images to automatically retrieve heights and winds of clouds, and other aerosol features above the ground using a stereoscopic method (Moroney et al., 2002a). For that, MISR needs a previous set of some fixed processing parameters, which will be applied equally to all scenes (Muller et al., 2002) to speed-up processing time. If all cameras measured high-resolution radiances, MISR data would be prohibitive. For this reason, only the nadir camera data and the red channels for the off-nadir cameras (12 of the 36 channels) are kept at the highest resolution (275 m), while data on the other channels are at 1.1 km resolution. This operational mode is the default and is called global mode (GM), useful for global studies of cloud heights and winds. However, MISR can also be configured to achieve high resolution for all the channels (36 channels) for a limited period of time and specific domains. This capability is known as the local mode (LM) and it is scheduled upon request from the user community.

The MISR operational product retrieves two types of stereo-heights. The zerowind heights assume that disparity in the same feature between camera views is

²https://eosweb.larc.nasa.gov/; last access 10/02/2019

³https://eosweb.larc.nasa.gov/sites/default/files/project/misr/guide/MISR_

Science_Data_Product_Guide.pdf; https://eosweb.larc.nasa.gov/project/misr/misr_ table; last access 14/02/2019

caused by parallax. Parallax is the difference in location of a projected feature on the ground due to different viewing angles (Moroney et al., 2002a). Smallest parallax is obtained from the nadir image (An) and is used as the reference camera. Wind-corrected heights are calculated after separating the contribution of the wind and the parallax to that disparity. They need the wind speed along-track and acrosstrack components to be computed and applied to the zero-wind cloud-top heights to produce wind-corrected heights at a horizontal resolution of 1.1 km. The wind direction and the along-track component of the wind speed are extracted using the Df/Da $(\pm 70^{\circ})$ and Bf/Ba $(\pm 46^{\circ})$ cameras imagery (Davies et al., 2007). Wind-corrected heights provide more accurate results, but are computationally expensive and at low spatial coverage, unlike the zero-wind heights, which offer excellent coverage (Kahn et al., 2007).

2.3 MODIS Instrument and products

The MODIS instrument is also aboard the NASA Terra satellite and observes the same scenes as MISR. MODIS detects from its far-infrared imagery, under cloud-free conditions, thermal anomalies at 1 km spatial resolution, named "fire pixels", (Figure 2.2). The detection method is based on an algorithm (Giglio et al., 2003) that exploits the strong emission of mid-infrared radiation from fires (Dozier, 1981, Matson and Dozier, 1981) and offers automated daily global fire information. In addition, MODIS provides estimates of the fire radiative power (FRP) for each fire pixel detected, a parameter used as a proxy of fire intensity. FRP is calculated from the differences in the radiance of each fire pixel and its background (Giglio et al., 2003). Two MODIS products are assimilated by MINX:

- 1. The Level 2 MOD14 Thermal Anomalies at a 1 km resolution, which includes FRP.
- 2. A global biome classification grid at 500 m of spatial resolution derived from

the MODIS Level 3 land cover product MCD12Q1 Land Cover Product⁴ (Friedl et al., 2010). This MCD12Q1 product classifies the land cover associated with each smoke plume in 17 International Geosphere-Biosphere Programme (IGBP) land cover classes and has an annual temporal resolution.



Figure 2.2: Natural-colour image collected by the Terra satellite across the Amazon on September 10, 2015. Actively burning areas, detected by MODIS's thermal bands, in red. https://www.nasa.gov/image-feature/goddard/wildfires-in-amazonian-region-of-brazil.

2.4 MINX Software

The MINX visualization and analysis interactive tool complements the MISR Level 2 operational stereo product for detailed studies of smoke, dust and volcanic ash.

⁴https://search.earthdata.nasa.gov/search?q=MCD12Q1%20V006; last access 14/02/2019

The MINX stereo-height algorithm was developed to overcome the limitations of the MISR operational product, as it enables the user to retrieve clouds, aerosols plume heights and winds at higher spatial resolution and better precision. Plumes are defined as regions of dense aerosol, with a well-defined discernible contour above the terrain and downwind of its source (Nelson et al., 2013), allowing to determine the direction of transport. Clouds, on the other hand, are not associated with any source and the direction of transport is not evident. MINX is written in the Interactive Data Language (IDL) and it can be downloaded from Github⁵, available for Mac OS X, MS Windows, and Linux platforms. Since the development of MINX, several versions have been released. The latest version is MINXv4, with substantial improvements that provide better quality smoke height retrievals. MINX has been used for the MISR Plume Height Climatology Projects (MPHCP)⁶. MPHCP aims at creating an aerosol injection height climatology to support wildfire, climate change, and air quality studies (Nelson et al., 2008b). In addition, MINX uses have been extended to many detailed studies of smoke plume heights over specific regions in the world (Val Martin et al., 2010, Mims et al., 2010, Tosca et al., 2011), studies of ash clouds from volcanic eruptions (Scollo et al., 2012, Kahn and Limbacher, 2012) and dust plumes from deserts (Kalashnikova and Kahn, 2008).

MINX interface allows the user to display the multi-angle nine camera images one by one or as an animated loop (Figure 2.3). This method enables the user to study plume and cloud dynamics, as it provides a similar 3D effect of the scene that could not be possible with a single image or multiple same-angle images. MINX requires all nine camera terrain-referenced imagery files (GRP_TERRAIN product) to derive accurate heights and winds over land (Jovanovic et al., 1998), and the geometric parameters product (GP_GMP), with zenith and azimuth viewing angles, which are both from MISR Level 1. An additional product is needed to perform stereo retrievals, the MISR Ancillary Geographic Product (AGP) at 1.1 km spatial

⁵https://github.com/nasa/MINX; last access 12/02/2019

⁶https://misr.jpl.nasa.gov/getData/accessData/MisrMinxPlumes2/; last access 09/02/2019

resolution, which contains the Digital Elevation Model (DEM) and surface feature IDs. Additionally, MISR Level 2 aerosol parameters (AS_AEROSOL) are used to obtain aerosol data, i.e., AOD, and TC_CLASSIFIERS to identify different types of aerosols (e.g., smoke or ash). All the cited products can be downloaded from the Earth data website⁷ after logging in. Table 2.1 summarises the MISR and MODIS products and files necessary to process smoke plume heights with MINX.



Figure 2.3: MISR nine camera views of a smoke plume on the 22nd of August 2010, in the Amazon

As the MISR operational product, MINX stereo-height algorithm provides zerowind and wind-corrected height values (Figure 2.4). MINX calculates the wind speed necessary to retrieve wind-corrected plume heights with an accuracy of 250 m by supplying the wind direction in the plume. For each grid in a plume, heights and winds are computed combining each of the six nearest camera neighbours to the nadir camera, used as a reference. Whenever the results of at least three camera pairs are similar, the retrieval is considered successful. In addition, if the MISR aerosol standard products are loaded in MINX, aerosol properties within the plume (e.g. Angstrom exponent, single-scattering albedo) will be extracted at 17.6 km of resolution.

⁷https://search.earthdata.nasa.gov/search

Product	Files	Description
MODIS MOD 14^{a}	MODIS/Terra Thermal Anomalies/Fire 5-Min L2 Swath 1km V005	fire pixels at 1 km and FRP in MW/pixel
$MI1B2T^{b}$	level 1 GRP_TERRAIN (terrain-referenced)	radiance files
$MIB2GEOP^b$	level 1 GP_GMP	camera and sun geometry
$MIANCAGP^b$	AGP – ancillary geographic	digital elevation data and surface type masks
$MIL2ASAE^{c}$	level 2 AS_AEROSOL	aerosol data: AOD, single-scatter albedo etc.
$MIL2TCCL^{a}$	level 2 TC_CLASSIFIERS	smoke/cloud mask files
antimal flog		

Table 2.1: Summary of MISR and MODIS files and products used to digitise smoke plumes with MINX.

"Optional files ^bRequired files ^cRequired only for plume studies

2.4.1 MINX Stereo retrieval algorithm

The retrieval process starts by matching images of camera pairs with the nadir camera and measuring the disparities within a plume. When the user determines the wind direction for each camera pair a height, wind-across-track and wind-alongtrack solution is achieved. This process is done for all camera pairs at the same point, and a maximum of eight heights and wind values are obtained depending on the number of camera pairs selected for matching and the number of successful retrievals. Then, the MINX stereo retrieval algorithm determines the more similar height and wind value among camera pairs for each point.

Height retrievals for static and in movement features

During Terra overpass, each MISR camera observes a feature in the atmosphere within seven-minute difference. The shift in the location of a feature between two cameras is its disparity, and consists of an along-track displacement parallel to the ground, and an across-track displacement in the orthogonal direction. The measure-



Figure 2.4: Comparison of wind-corrected and zero-wind stereo-height pixels per plume in a smoke plume climatology over North America. (Adapted from Val Martin et al. (2010))

ments of these two components provide the primary information to compute stereo heights and winds (Moroney et al., 2002a).

In the case of static features in the atmosphere, the along and across-track disparities due to motion are zero, the across-track disparity due to parallax is zero and the along-track disparity is only due to parallax. Therefore, its height can be determined by knowing this parallax disparity and the angle of the non-nadir camera (Moroney et al., 2002b)(Figure 2.5). This is called the zero-wind height. The zero-wind height can also be performed for features in movement however, errors range from tens of meters to kilometres, depending on the height of the feature, the wind and the camera pair used.

If the feature is not stationary, then the height, the wind speed in the along- and across-track directions are unknown for a camera pair, assuming no vertical motion. In the MISR operational product this is solved by adding a third camera pair (D) and making some assumptions (Zong et al., 2002) (Section 2.2). In MINX, there are two cases to perform the stereo height retrieval. The case in which the feature moves only in the across-track wind component and the case in which the feature moves in both components, along and across-track. In the first case, the along-track disparity due to motion is zero and the height is determined in the same way as the zero-wind height performance, assuming that the along-track disparity is only due to parallax. The across-track wind speed can then be determined by converting the across-track disparity to map distance and dividing by the time between the two camera viewing angles. To compute this height the Earth's curvature, terrain height and other factors need to be considered. When a feature is moving in the two components, the across-track wind component will be determined using the method described above but the along-track component includes the contribution of the parallax and the real displacement due to the along-track wind. In this case, the height and the along-track wind components need to be determined, knowing only the along-track disparity.

The MINX stereo-height retrieval algorithm allows the user to provide the wind



Figure 2.5: MISR view with respect to features in the atmosphere, disparities produced by parallax and due to motion of the MISR instrument and the feature. (Image adapted from https://github.com/nasa/MINX/blob/master/webdoc/MINX_Doc5.pdf).

direction when digitising (Section 2.4.2). For any point in the user-supplied wind direction, the wind direction can be calculated as the slope of the digitised line, which is the ratio of the along- and across-track distances and the ratio of the wind speed in the along- and across-track components. If one of the wind speeds is known, the other can be calculated from the slope of the line and the along-track wind, and height can be determined at high resolution using a camera pair. This stereo retrieval method is applied to each point of the digitised plume using all camera pairs and the nadir camera as reference.

Image matching

The image matching process consists of finding a feature in a non-nadir image that corresponds to that feature in the nadir image and measuring its disparity. During the MINX stereo height retrievals, all cameras selected for image matching should be paired with the nadir camera. Meaning that whenever a feature is not visible in the nadir camera but visible in any of the off-nadir cameras, MINX will fail at performing the stereo retrieval. This process uses a square template-based reference image in the red-band, centred on the pixel of study in the nadir view. The match is performed when a target pixel in the comparison image is found to best correlate to the reference template. This method provides more accurate matcher results applied to features that extend through the template than to a single pixel at the centre. The image matching process requires intense use of the CPU, and processing times depend on the hardware and the area to perform the match. Larger templates require longer processing times and can improve the retrieval coverage, but the smaller are usually more successful for fine spatial detail when plumes have small variations in height. The matcher template size is defined by the user. In the case of the Amazon climatology of smoke plumes, the default option (medium size) was chosen, which provides enough detail at reasonable processing times.

Determination of height and wind

Once the successful matches for the camera pairs produce the retrieved results for a sample point, these results are then evaluated to determine a consensus height and wind for all camera pairs matching. The mean heights and winds are calculated for those camera pairs retrievals that are more similar to the median values, used to soften the effects of the outliers. Heights and wind results are discarded if they do not fall into a threshold distance from the media, where the threshold distances are calculated dynamically.

Spectral band

The red band is the high-resolution band in the global mode of the MISR operational product because of its larger contrast between atmospheric features over ocean and land (Diner et al., 1998). However, over bright surfaces like grasslands or deserts, or in the case of low-dense features stereo-height retrievals in red band is not preferred (Mims et al., 2010). Increases in wavelength lead to decreases on atmospheric scattering and smoke will be more transparent, allowing to see the terrain through it. If in the reference image the terrain is seen through the smoke plume, the image matching will be more difficult to process, performing less successful height retrievals. Therefore, blue band retrievals generally offer greater sensitivity to thin aerosol layers and over bright surfaces.

Before the release of MINXv4 the default spectral band option was red. However, the user could select the blue band if applicable to the characteristics of the plume and background. This should be configured by the user manually at the start of digitising each plume, which entails additional time into the digitising process. Since MINXv4, two plume height retrievals are performed for each plume. One retrieval using red-band data and the other using blue-band data. Each retrieval is treated as a different plume, but they share the same aerosol properties, from the MISR aerosol product, and the same plume coded name in exception of a letter, "R" or "B", depending on the spectral band ("R" for red and "B" for blue band). This new capability allows the user to choose between the best quality height retrieval plume, but it doubles the number of plumes created.

2.4.2 Digitalization of smoke plumes with MINX

MINX can be used to digitise and study a single plume or to create a large climatology of smoke plumes observed by MISR. The files generated by MINX include among other parameters the location and time of the plume, different statistics for smoke plume heights based on the individual height retrievals, the radiative power of the associated fires, the direction of transport of plumes and aerosol properties.

MISR files are large (~ 2 Gbytes/orbit) and tedious to download for projects that cover large periods of time or areas, like this study. The MINX "Plume Utilities" is a tool designed to limit the amount of MISR data to download and process, reducing considerably processing time and computing space. This tool allows the user to select only the MISR orbits and blocks where it is likely to find smoke plumes, rather than download and visually inspect all MISR images for the time range and area of study. In the case of smoke plumes from wildfires, MINX uses the MODIS Terra thermal anomalies product, as mentioned in Section 2.3. By loading the fire pixels, MINX generates a list of the MISR orbits and blocks with coincident active fire pixels. To do so, the user needs to provide the geographic bounds and the time range for the study when ordering the MODIS MOD14 thermal anomalies. The "Plume Utilities" tool reduces the number of MISR files to download by a factor of 100 or more (Nelson et al., 2008b). In addition, the MODIS thermal anomalies files are read by MINX and displayed as a layer of red dots on MISR imagery (Figure 2.2), which helps the user identify plumes, and allows MINX to compute the approximate total FRP for each plume.

Once the necessary MODIS fire pixels and MISR Level 1 and 2 files are downloaded (Table 2.1), MINX is ready to process them, display and compute stereo heights of smoke plumes. Before digitising, the user needs to load and link the MODIS fire pixels to the MISR orbits and paths images. This step includes specifying some parameters as the minimum number of fire pixels to consider or their confidence level. For the specific case of the Amazon climatology, the default options were selected as presented in Figure 2.6. Following this, MINX loads the MODIS thermal anomalies on the MISR images (Figure 2.2). At this point, the user inspects the MISR multi-imagery block by block with the coincident fire pixels superimposed and identifies smoke plumes. The ability to visualise each plume in nearly 3D is decisive to study its structure and dynamics.

The digitising process starts by drawing with the mouse the contour of the plume, starting at the fire source, and the direction of transport. Plumes direction can be digitised with as many points as necessary, being common to draw only two points in the case of a quite linear plume. It is important to make sure that all fire pixels associated with the same plume are contained within the digitised area, as MINX computes the total FRP for each plume and if any fire pixel is not included, it



Figure 2.6: MODIS fire pixel parameters to configure when loading in MINX

will not be summed to the total. Once the contour and direction of transport in a plume are digitised, the MINX stereo-height retrieval algorithm automatically starts, and the drawn plume area is filled in with colour-coded pixels, where colours correspond to the magnitude of the MINX-computed wind-corrected heights. This is achieved only if stereo-heights and winds are successfully retrieved. Figure 2.7 shows the digitising process of a plume observed on the 22nd of August 2010 in the Amazon (O56797-B99-SPWR11). On the left panel, the MINX nadir camera image is displayed with the overlayed red dots representing active fire pixels from the MODIS thermal anomalies product, and the mouse-digitised contour (blue) and wind directions (yellow) of the plume. On the right panel, the retrieved windcorrected smoke plume heights are displayed on the MISR image in a colour scale, where blue represents low and red high smoke plume heights.



Figure 2.7: MISR nadir image of a smoke plume on the 22nd of August 2010 in the Amazon and MINX smoke wind-corrected height retrievals computed within the plume.

2.4.3 Summary of MINX outputs

MINX automatically generates some outputs for each plume successfully digitised and provides its location in the MISR orbit (Figure 2.8). Firstly, MINX generates a plume name which will be unique and common to all the output files associated with that plume. The name convection follows some rules that are briefly explained below. To illustrate the outputs, Plume O56797-B99-SPWR11, in Figure 2.7, is used as example.

- 56797 is the orbit number and correspond to the 2-7 characters in the name. The minimum value is 000995 and the maximum 9999999.
- 99 is the block number where the first point was digitised and corresponds to the 11-12 characters. The minimum value is 001 and the maximum 180.
- S is the 14th character and defines the type of aerosol. S is for smoke, D is for dust, V is for volcanic ash and W is for water (clouds/snow).
- P is the 15th character and defines the geometry of the digitised feature. Being
 P for polygon and L for line.
- W is the 16th character and is used to define if the direction of the wind was provided or not. Therefore, W will be assigned to plumes, where wind



Figure 2.8: Location of the Plume O56797-B99-SPWR11 in the MISR orbit and path. Block 99 in the orbit 56797 is shown with a red square

direction is digitised and zero-wind heights, wind speeds and wind-corrected heights are retrieved. On the contrary, when no wind direction is supplied the letter N is assigned. This is the case of clouds.

• R defines the band used in the height retrieval. It is the 17th character and it can be either R or B, for red or blue band, respectively.

• 11 corresponds to the last characters (18-19) and it refers to the sequenced number of plumes digitised for that block. The minimum value is 01 and the maximum 99.

Immediately after digitising a smoke plume MINX generates a series of outputs that are displayed automatically in the screen. These outputs include:

• Height Profiles

The height profiles are plots where the x-axis represents the distance from the first point digitised, usually coincident with the MODIS fire source pixel, and the y-axis represents the height in meters above sea level of each point successfully retrieved and coloured in red (zero-wind) and blue (wind-corrected) (Figure 2.9). Terrain elevation is also represented for each point (green colour). It is typical to find more zero-wind corrected points than wind-corrected points (Figure 2.4, Section 2.4). Stereo-height retrieval profiles display more data whenever the number of successfully retrieved heights is increased and the area of the digitised plume is large, as many points will be at the same distance from the source. However, there are situations in which due to the characteristics of the plume (generally low dense plumes) the number of successful retrievals is low and only a few points will be represented in the profile. This is reflected in the quality of the plume, determined by MINX. In the case of a feature with no apparent direction or movement (smoke clouds), only zero-wind heights are represented. For plumes digitised in both blue and red bands (only MINXv4), MINX creates two different profiles, one for each plume/band.

• Wind Profiles

As in the case of the heights profile, wind-corrected profiles are represented by the successful retrieval points from the source fire pixel. Points are split into the across-track and the along-track components of the wind represented in green and red colours, respectively. Wind speed is positive if directed towards the north and east (top and right directions on MISR images). If the along-track component of wind is directed toward the north, then the wind-corrected height will be less than the zero-wind height. Wind retrievals and wind-corrected heights are less reliable when the wind direction is more aligned to the MISR along-track direction because the along-track motion of the satellite becomes more difficult to separate from the plume motion. MINX determines a quality flag of the retrieval taking this into account. Figure 2.9 shows the MINX vertical height and wind profiles for the same plume on the 22nd of August 2010 (O56797-B99-SPWR11). For this example, the windcorrected height increases from about 2 km above sea level (asl) in the first 40 km from the origin to a maximum height of ~4 km. Median smoke plume height is around 3 km. The along-track wind component shows high variability from the first 40 km, within a range of -18 to -1 m/s, whilst the across-track ranges from -6 to -2 m/s. The sum of these speeds yields around -6 m/s.

• Aerosol Histograms

The MISR standard aerosol product provides aerosol data at 17.6 km of resolution whenever the optical depth is lower than 2.0, free of cloud contamination and the terrain is not abrupt. MINX extracts aerosol data from MISR however, results in a single small entire plume may be the same for the whole plume due to the coarse aerosol product pixel. The aerosols histograms created by MINX include optical depth, single-scatter albedo and the tau fraction by particle-type (Figure 2.10). Both optical depth and single-scatter albedo contain spectral data, represented by the colours of the bars. The tau fraction by particle-type histogram represents the fraction of the green-band optical depth value that corresponds to small, medium and large particle sizes and to spherical particles.

In addition to these outputs, for each plume, MINX processes and stores detailed information of each retrieved point in an ASCII file (Figure 2.11). The header of this file contains general information about the plume, i.e., the orbit, path and



Figure 2.9: Height and wind profiles for the Plume O56797-B99-SPWR11.

block number, date acquired, UTC time, MINX version, user name, date digitised, plume name, aerosol and geometry type, band of retrieval, first point latitude, first point longitude, perimeter, area, mean terrain elevation, retrieval quality, biome (only in MINXv4), MISR product files loaded in MINX, etc (Figure 2.11, top left panel). Following the header, there are three tables, the first two tables contain the coordinates of the points that define the area of the digitised polygon and wind direction (Figure 2.11, top right panel), and the third table contains one data point



Figure 2.10: Aerosol histograms for the plume O56797-B99-SPWR11 on the 22nd of August 2010.

per row and several columns of parameters (Figure 2.11, bottom panel), where each data point corresponds to the location where MINX retrieves data. Some of the information contained in the file is presented in Figure 2.11 and the most relevant are defined below.

- Retrieved with matcher: specifies the size of the image matcher used in the retrieval.
- Retrieved with cameras: specifies the set of MISR cameras used in the retrieval.
- Power of fire: is the cumulative fire radiative power (FRP) in MW, for all the MODIS fire pixels contained in the digitised polygon that represents a plume.
- Best median and top height: these parameters provide the best estimates for the median and the maximum height, in meters above sea level, in which outliers are removed following different smoothing techniques (Kahn et al.,

prbit number : 56/97 Path number : 229 Block number : 22010-08-222 UTC time : 2410-98-22 UTC time : 141:09:54 MINK version : 34.0 User name : Laura	POLYGON:	22 poi Long- itude -57.802 -57.843 -57.938	nts in th Lat- itude -10.607 -10.619 -10.672	is table of Blk Samp 99 1551 99 1535 99 1499	Jefine th Line 182 188 212	ne digit	ized bou	nding poly	gon if pr	esent.
Date digitized : 2014-12-23 Region name : : 056797-699-SPWR11 Region aerosol type : Smoke Region genometry type : Polygon Region wind dir type : Direction provided Retrieved with band : Red Match blue in An only? : No Retrieved with matcher : Kedium Retrieved mith matcher : Kedium Retrieved mith matcher : Kedium Retrieved mith matcher : Kedium	4 5 6 7 8 9 10 11 12 13 14 15 16	-57.997 -58.070 -58.156 -58.255 -58.373 -58.489 -58.411 -58.331 -58.223 -58.173 -58.111 -58.131 -58.131 -58.066	-10.725 -10.757 -10.818 -10.900 -10.988 -11.047 -11.180 -11.286 -11.269 -11.208 -11.208 -11.084 -10.984 -10.9867	99 1477 99 1449 99 1417 99 1380 99 1336 99 1292 99 1327 99 1362 99 1404 99 1425 99 1445 99 1445 99 1445	235 250 277 313 352 379 430 470 460 434 427 383 346 294					
First point laftude : -57.80.67 First point laftude : -10.60747 Perimeter length (km) : 228 Area (sq km) : 1633	18 19 20 21 22	-58.028 -57.940 -57.880 -57.825 -57.802	-10.827 -10.774 -10.704 -10.648 -10.607	99 1468 99 1501 99 1523 99 1543 99 1551	277 253 223 199 182					
Area per point (sq km) : 1.210 Wind-corrected points : 967 Best median ht (m ASL) : 2801 Best top ht (m ASL) : 2801 Best top ht (m ASL) : 3864 StdDev metric, corpht : 242 [whdDir=AlongDir](deg) : 33 Power of fire in NW : 3336.7 Retrieval quality : GGOOD Level 1 radiance file : MISR_AMI_GRP_TERRAIN_GM_P229_0056797_AN_F03_0024.hdf Cam/Sun Geometry file : MISR_AMI_AGP_P229_L01_24.hdf Cam/Sun Geometry file : MISR_AMI_AGP_P229_L0256797_F03_0013.hdf Aerosol product file : MISR_AMI_AS_AEROSOL_P229_0056797_F07_0012.hdf	DIRECTIC Pt# 1 2 3 4 5 6 7 8 9 10 11 12 13	DN: 13 p itude -57.845 -57.845 -57.889 -57.983 -57.985 -58.032 -58.032 -58.139 -58.139 -58.174 -58.215 -58.280 -58.350	boints in Lat- itude -10.644 -10.680 -10.710 -10.761 -10.850 -10.856 -10.929 -10.996 -11.068 -11.152 -11.217	this table Blk Samp 99 1551 99 1535 99 1535 99 1535 99 1545 99 1483 99 1465 99 1445 99 1445 99 1445 99 1427 99 1428 99 1378 99 1352	2 define Line 182 198 214 227 249 262 271 291 349 379 349 379 443	the dig	itized d	irection 1	ine if pr	esent.
RESULTS: 1290 points in this table are samples where Nownd heights o Long- Lat- Km to Dg Gw Terr Featu Optical Depth by Band Single-scattering Albedo Tau Fraction Pt# itude itude Blk Samp Line Pt 1 Rel N Elev Nownd Blue Green Red NIR Blue Green Red NIR Small Mediu	r fire re Ht (by Par W/Wnd m Lar	power w (m) rticle 1 Fltrd rge Sp	vere ret Windsp Type Across Dher E	trieved. Deed (m/ Ang Po Along T Exp Mw	's) wer R 'otal Vatt (tefl Blue)->1	Albedo BT21 Greer Deg K	by Ban BT31 Red Deg K	d BBT21 NIR Deg K	BB TO BBT31 Albeo Deg K
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	-99 1 0.0 -99 9 0.1 -99 9 0.1 -99 9 0.1 -99 9 -9.9 9 -9.9 0 -99	$ \begin{array}{ccc} -99 \\ -99 \\ 197 \\ 0. \\ -99 \\ 197 \\ 0. \\ -99 \\ 197 \\ 0. \\ -99 \\ 197 \\ 0. \\ -99 \\ -99 \\ -99 \\ 0. \\ -99 \\ 0. $	-99.9 -99.9 600 1 -99.9 600 1 -99.9 600 1 -99.9 600 1 -99.9 -99.9 -99.9	-99.9 -9 .959 -9 .206 -9 .206 -9 .206 -9 .206 -9 .99.9 -9 .206 -9 .99.9 -9 .447 -9 .99.9 -9 .447 -9	19.9 19.9 19.9 19.9 19.9 19.9 19.9 19.9	0.30 1.999 - 0.26 1.999 - 0.26 1.999 - 0.26 1.999 - 0.26 0.26 0.26 0.26 0.26 0.26 0.26 0.26 0.299 - 0.26 0.20 0.2	0.24 -999.9 0.20 -999.9 0.21 -999.9 0.20 -999.9 0.21 -999.9 0.21 -999.9 0.21	0.21 -999.9 0.18 -999.9 0.17 -999.9 0.18 -999.9 0.18 -999.9 0.18 -999.9 0.20	0,25 -999.9 0,28 -999.9 0,29 -999.9 0,28 -999.9 0,28 -999.9 0,28 -999.9 0,28	0.19 -999.9 0.19 -999.9 0.19 -999.9 0.19 -999.9 0.19 -999.9 0.19 -999.9

Figure 2.11: MINX ascii file for the plume O56797-B99-SPWR11, observed on the 22nd of August 2010.

2007, Nelson et al., 2013).

Data quality: specifies the quality of the plume height retrievals based on the number of heights successfully retrieved in a plume, the percentage of the plume's area filled with successful heights, the standard deviation of the mean of the successful heights and the wind direction. This flag can adopt three values, "GOOD", "FAIR" or "POOR", and no value, when the number of retrieved points with wind-corrected heights is too small. Only smoke plumes classified as "FAIR" or "GOOD" should be included in studies of smoke plumes. However, this selection does not guarantee that the windcorrected heights have the highest accuracy. If the plume was digitised with a wrong wind direction, then the retrieved heights will be less accurate. For the Amazon climatology, 51% of the plumes had low quality and were discarded from the original dataset. The climatology of smoke plumes over the Amazon consists of 1.21 TB (75232 files and 474 folders), including the required files from MISR and MODIS. In total, 10858 plumes were digitised with different versions of MINX (v2–4), of which only 5393 plumes were included in Gonzalez-Alonso et al. (2019) (Chapter 3). The climatology contains eight years of smoke plumes observed during the burning seasons (July-November), from 2005 to 2012, across the Amazon domain, defined by the coordinates 80°W–40°W of longitude and 25°S–5°N latitude.

The inspection of the plumes via animation and the digitising process in MINX, although both are quite straightforward, are time-consuming and memory and CPUintensive. Furthermore, the time required to read MISR files is proportional to the number of MISR blocks selected to load. Loading all nine camera images for only five blocks of MISR data over a fast network (there are usually 144 blocks of valid data per MISR orbit) requires 35 seconds on a 2.2 gigahertz windows 7 system with Intel Core i7 processor and 4 gigabytes of memory (Nelson et al., 2008a).

2.4.4 MINX Additional tools

MINX offers other utilities, which can be very useful depending on the project objectives and the degree of detail required in the study. Some of these utilities have been applied in the development of the smoke plume height database over the Amazon (Chapter 3). The most commonly used are:

I Camera registration correction

Before image matching is performed, images for each camera pair must be geometrically registered to improve stereo height retrieval accuracy. This is achieved by reducing errors in camera-to-camera geometric registration (Davies et al., 2007). Errors in one pixel (275 m) of co-registration can entail differences in the range of 160 m to 560 m of the height retrievals (Zong et al., 2002), depending on the cameras pair selected for matching, but mean geometric coregistration error for all cameras is less than 275 m (Jovanovic et al., 2007). These errors must be avoided before digitising. For that, the user can superimpose a "fixed grid" over the MISR imagery and focus on a fixed feature on the terrain (Figure 2.12). If the feature is displaced with respect to the fixed grid when displaying the nine angle-views, correction should be applied. Registration errors due to failures in the Digital Elevation Model (DEM) from the MISR Ancillary Geographic Product (AGP) files cannot be corrected.



Figure 2.12: MINX camera registration correction. Fixed grid in yellow is displayed superimposed on MISR imagery

II digitising options

When the user starts to digitise a dialog box pops at the MINX interface where the user can select different options according to the study and desired results (Figure 2.14). First, the type of aerosol for which height retrievals will be processed, either smoke, dust, ash or water/ice. Then, the user needs to choose other options, i.e., a line or polygon to retrieve heights, provide wind direction or not (for clouds), select camera pairs for matching, etc. In the case of the Amazonian climatology, the setting was configured for smoke polygons with wind direction and default MINX options (Figure 2.14 depicts the setting selection for the Amazon smoke plumes). Depending on the user's selection, the digitised feature will have coded letters allocated for those specifications and they will be part of the digitised feature name (Section 2.4.3).



Figure 2.13: Dialog box to set the parameters to digitise a feature in MINX.

III Masks

There are four support vector machine (SVM) masks: cloud, smoke, land and dust masks, which are only available if the MISR standard Level 2 TC_CLASSI-FIERS product is loaded at digitising (Table 2.1). Masks are only visible if the user presses the button in the MINX interface (Figure 2.14). The SVM masks provide within a confidence level the probability of each pixel to be or not to be that mask type and assist the user to identify features as a specific type (e.g. cloud or smoke).

IV Re-digitise, Delete

MINX offers the possibility to re-digitise or delete plumes already digitised whenever retrievals are not satisfactory or when after inspecting the height and wind profiles, better results are possible. This capability is very useful as sometimes it is complex to understand the dynamic of the smoke and trying different shapes, reducing the area digitised or changing the wind direction, can lead to more successful retrievals.



Figure 2.14: Cloud mask tool with black (no value), dark red (high confidence pixel is not mask type), light red (low confidence pixel is not mask type), light green (low confidence pixel is mask type) and dark green (high confidence pixel is mask type).

2.4.5 Interpreting wind direction

Determining the wind direction in the process of digitising is one of the most important tasks for the user. Although it can look trivial, wrong determination of the wind direction can be translated in errors of several kilometres in the windcorrected heights for fast moving plumes. The direction of motion can usually be determined by investigating the scene, i.e., see the direction of shadows over the terrain or observe the direction of the contiguous plumes. However, there are some cases in which the direction is not so obvious and detailed inspection of the plumes and scenes is required. This is the case for low dense plumes over bright surfaces with not well-defined contour, plumes with a column of air ascending very fast in the vertical, where the convection at the fire is stronger than winds advection, or smoke plumes with two directions, where the plume seems to bifurcate.

2.4.6 Other applications of MINX

In addition to smoke plumes, MINX can be used to study volcanic and dust plumes, as well as hurricanes and boundary layer clouds. For example, Scollo et al. (2012) used MISR multi-year observations from explosive events at Mt. Etna and assessed the height of the volcanic aerosol plume with MINX stereo matching technique. They also used MISR aerosol product to distinguish between the sulphate and/or water vapour dominated volcanic plumes and those dominated by ash, indicative of eruption strength to constrain some parameters in volcanic ash dispersion models. Figure 2.15 represents the 2001 Flank Eruption, on the 22nd of July from Scollo et al. (2012). The explosive activity originated from two craters at 2550 m asl and the eruption extended 24 days, from the 17th of July to the 9th of August, 2001 and it is recorded as one of the most unusual and complex eruptions of Mt. Etna in the last 300 years⁸, causing 3.1 USD million damage, including losses in tourism and agriculture⁹. The ash plumes were captured by MISR on the 20, 22, and 29th of July, and using MINX, wind-corrected heights were determined. In particular, in Figure 2.15 the ash plume was dispersed toward the SE and reached ~ 6 km of altitude.

MINX has also been used to characterise the transport and aerosol optical depth of dust plumes across different regions (Kalashnikova and Kahn, 2008, Flower and Kahn, 2017, Li and Sokolik, 2018). An example of MINX dust height retrievals is depicted in Figure 2.16, along the Red Sea coast of Sudan (Nelson et al., 2013), on the 24th of July 2010, where strong onshore winds (20 m/s) lofted dust from the dry riverbeds in the Red Sea to an altitude of 1.4 km. Maximum wind-corrected heights and wind speed are observed within 5–10 km and 100 km, respectively, downwind from the source.

⁸http://www.ct.ingv.it/en/11-notizie/news/

⁴⁰⁶⁻¹⁰⁻years-ago-the-july-august-2001-eruption-of-etna.html; last access 27/02/2019 ⁹https://earth.esa.int/web/earth-watching/natural-disasters/volcanoes/

content/-/asset_publisher/NBxzsX9lcE11/content/etna-volcano-italy-2001; last access
27/02/2019



Figure 2.15: The 22nd July 2001, eruption of Mt. Etna in Italy on orbit 8476, block 61. Image from the MISR nadir view with the wind-corrected heights retrieved by MINX (top left panel), wind-corrected height profile (gray points) (bottom panel), (both adapted from Scollo et al. (2012)) and location of the block and MISR orbit (in red circle; top left panel)



Figure 2.16: Dust over the Red Sea on MISR orbit 56371, block 75–76, from 24 July 2010: (a) MISR nadir image; (b) MINX wind-corrected heights over the MISR nadir image; (c) MINX height profile; (d) MINX wind speed profile. (Image adapted from Nelson et al. (2013)). In this case, the plume descends as it approaches the sea, as the Marine Atmospheric Boundary Layer (MABL) is shallower.

2.5 MINX Limitations and biases

Despite being a powerful software to study plume dynamics, there are some limitations in MINX that need to be taken into account to avoid errors and biases during the digitising process (Nelson et al., 2013). Errors and biases considered during the development of the smoke plume climatology over the Amazon are summarised below:

- Low quality of height retrievals due to low optical depth aerosols: MINX stereo height retrieval algorithm needs first to identify features common to the multiple cameras to perform a cross-correlation between pairs of camera images. If the feature is not texturally discernible from the background, then the algorithm performs the matching with the surface and only zero-wind heights close to the ground are computed. This is typical of low-dense plumes, where the terrain surface can be seen through the smoke, enhanced over high terrain reflectivity, like grassland or thin forests. In particular, for the Amazon climatology, this bias does not have a big impact because the retrieval quality for low dense smoke plumes was poor and therefore, those plumes were discarded.
- Low quality of height retrievals for homogeneous aerosols: MINX fails at retrieving both zero-wind and wind-corrected heights, whenever the smoke plumes or clouds are texturally homogeneous. This bias should be considered specifically for clouds, usually more homogeneous than smoke plumes.
- Contamination by water clouds: MINX users need to pay attention to the presence of clouds in a scene. If clouds are included in the digitised area, height retrievals will be biased high. To avoid this bias, MINX offers the mask tool from the MISR aerosol product, which allows the user to overlay a cloud mask layer to show the confidence to which aerosols in the image pixels are water clouds or not. However, if the user is not sure if a cloud is present in the scene, it is better to avoid digitising it. Over the Amazon, the presence of clouds is quite common, especially during the wet season and plumes were

digitised under cloud-free conditions whenever applicable, avoiding to include them in the digitised area.

- Digitising margins of smoke plumes: In the periphery of the plumes there is a potential bias towards lower quality or lower heights because smoke is more diffuse. For this reason, it is convenient to reduce the area of the plume, excluding the margins. Plumes over the Amazon are usually small and reducing the digitised plume area leads sometimes in a lack of successful retrievals. However, the conservative approach was followed and the area of the plume was reduced to the core as much as possible.
- Pyro-cumulus clouds associated with plumes: Pyro-cumulus constitute a type of clouds generated by intense fires associated with large buoyancy above the fire, which can reach substantially high altitudes. Digitising pyro-cumulus entails a bias towards higher smoke plume heights and they should be excluded at digitising. However, excluding them may translate in a decrease in the number of smoke plumes above the boundary layer. Over the Amazon, pyro-cumulus are not common, but they were excluded whenever present.
- Incorrect wind direction: If the user provides a wrong direction of the plume, motion errors in the wind-corrected height retrievals occur and the quality of the retrieval can be affected. This is a common problem, as sometimes the direction is not evident as mentioned in section 2.4.5. For this reason, inspecting the scene with caution is required and it is recommended to digitise the plume providing multiple solutions to the wind direction to finally select the best result for a plume. This strategy was followed when constructing the Amazon smoke plume data.
- Plume wind direction along-track: If the plume direction is parallel to the motion of MISR (along-track), MINX fails at separating the component of motion due to the wind from parallax error and it translates in low-quality retrieval. This is considered by MINX in the quality flag and plumes with low

quality were excluded from the Amazon climatology.

An additional bias arises due to the use of different versions of MINX. As mentioned before, the climatology of smoke plumes across the Amazon was created with different versions of MINX(v2-4). Plumes digitised with versions earlier than v4 had the red-band retrieval as default. The number of successful retrievals in red band is lower compared to blue band, particularly over bright surfaces and for low optical depth smoke plumes, as it is usually the case over the Amazon. Since MINXv4 all plumes are digitised in dual-band and MINXv4 determines the best option for each plume. This functionality allows the user to choose the best quality plume between both options, translating into a larger number of successfully retrieved plumes. For the climatology of Amazonian smoke plumes, blue-band plume retrievals were 60% more successful compared to 36% for the red-band retrievals.

Chapter 3

Biomass burning smoke heights over the Amazon observed from space[†]

3.1 Introduction

Fires burn across the Amazon region every year, releasing large amounts of trace gases and aerosols into the atmosphere (e.g., Andreae and Merlet, 2001). The majority of these fires are of anthropogenic origin: for deforestation, preparation of agriculture fields, conversion of cropland to pasture or road and city expansion (Cochrane, 2003). Between 1976 and 2010, deforestation fires destroyed more than 15% of the original Amazonian forest (Aragao et al., 2014). Most of these fires burn in the so-called arc of deforestation, along the eastern and southern borders of the Amazon forest, during the dry season (typically from July to November) (Malhi et al., 2008). However, significant variability exists, caused by changes in meteorology, drought

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and land-management policies (e.g., Nepstad et al., 2006, Van der Werf et al., 2010, Alencar et al., 2011). Amazon fires can contribute up to about 15% of the total global biomass burning emissions (Van der Werf et al., 2010). These emissions have important implications for air quality, atmospheric composition, climate and ecosystem health (e.g., Ramanathan et al., 2001, Johnston et al., 2012, Pacifico et al., 2015). For example, air pollution from deforestation fires is estimated to cause on average about 3,000 premature deaths per year across South America (Reddington et al., 2015) and may decrease the net primary productivity in the Amazon forest as a result of increases in surface ozone (Pacifico et al., 2015).

Fires are also an important source of buoyancy locally, which in combination with other atmospheric properties determines the vertical distribution of fire emissions in the atmosphere near the fire source (i.e., injection height). The altitude to which smoke is injected is critical, as it determines the lifetime of the pollutant, its downwind transport dispersion pathway, and the magnitude of its environmental impact (e.g., Jian and Fu, 2014, Archer-Nicholls et al., 2015, Paugam et al., 2016, Zhu et al., 2018). Space-borne observations have been used to study smoke injection heights across the world. Using Multi-angle Imaging Spectro Radiometer (MISR) stereoheight retrievals, smoke plume heights have been assessed across North America (Kahn et al., 2008, Val Martin et al., 2010), Indonesia (Tosca et al., 2011), Australia (Mims et al., 2010), southeast Asia (Jian and Fu, 2014), and Europe (Sofiev et al., 2013). For example, Val Martin et al. (2010), using a 5-year climatology of smoke fire plumes and smoke clouds observed by MISR across North America, showed that wildfire smoke can reach altitudes from a few hundred meters above the ground to about 5 km, and that 5-30% of the smoke plumes are injected into the free troposphere (FT), depending on the biome and year. Related work also demonstrated the important effect that fire radiative power, i.e., a proxy of fire intensity, and atmospheric conditions have on the initial rise of fire emissions (Freitas et al., 2007, Kahn et al., 2007, Val Martin et al., 2012). Tosca et al. (2011) reported that less than 4% of smoke plumes reach the free troposphere, based on a MISR 8-year climatology from tropical forest and peatland fires over Borneo and Sumatra, and found that the greatest plume heights were recorded during an El Niño year over Borneo.

Smoke plume heights have also been determined using space-borne lidar observations from CALIOP (Labonne et al., 2007, Huang et al., 2015), aerosol index from the TOMS and OMI instruments (Guan et al., 2010), and CO observations from TES and MLS (Gonzi and Palmer, 2010). Huang et al. (2015) used a multi-year record of CALIOP vertical aerosol distributions to study smoke and dust layer heights over six high-aerosol-loading regions across the globe. Specifically over the Amazon, they found that on a broad scale, smoke layers are typically located above boundary layer clouds, at altitudes of 1.6–2.5 km. Consistent with the smoke altitudes detected by CALIOP, an analysis of injection heights using CO observations from TES and MLS estimated that about 17% of fire plumes over South America reached the free troposphere in 2006 (Gonzi and Palmer, 2010).

Numerous studies have sought to understand the impact of biomass burning in the Amazon on local to hemispheric scales. In particular, during the past decade, several aircraft campaigns have been designed to study the effect of biomass burning on greenhouse gases, aerosols loading, clouds, regional weather and/or climate over the Amazon [e.g., BARCA (Andreae et al., 2012), SAMBBA (Allan et al., 2014) and GoAmazon (Martin et al., 2016)]. For example, modelling studies during SAMBBA showed the importance of the vertical representation of aerosols from biomass burning over the region (Archer-Nicholls et al., 2015), as biomass burning can modify local weather (Kolusu et al., 2015) and regional climate (Thornhill et al., 2017). Based on lidar observations taken in six research flights during SAMBBA (September 16–29, 2014), Marenco et al. (2016) reported the presence of two distinct smoke aerosol layers, a fresh smoke layer extending from the surface to an altitude of 1–1.5 km, and an elevated and persistent layer of aged smoke at 4–6 km. During the 2008 dry biomass season, continuous raman lidar measurements of optical properties taken in Manaus (2.5°S, 60°W) also detected biomass burning layers at 3–5 km heights, although most of smoke was confined below 2 km (Baars

et al., 2012). Whilst the results from these aircraft and in-situ lidar observations are significant, there are no analyses yet that seek to quantify the long-term average vertical distribution of smoke from fires across the Amazon, and to identify the key factors that control plume rise over this region.

We present here an 8-year climatology of smoke plume heights over the Amazon, derived from observations by the MISR and CALIOP instruments on board the NASA Terra and CALIPSO satellites, respectively. These data are analysed in combination with measurements of Fire Radiative Power (FRP) from NASA MODerate resolution Imaging Spectroradiometer (MODIS) instruments, assimilated meteorological data from MERRA-2 and drought condition indicators from the MODIS Drought Severity Index (DSI). The objectives of this work are to characterise the magnitude and variability of smoke heights from biomass burning across the Amazon, and to assess the influence of biome type, fire intensity, local atmospheric conditions, and regional drought on smoke vertical distribution as well as aerosol loading.

3.2 Data and Methods

We use a combination of remote sensing data from multiple sources to build a comprehensive climatology of smoke plume heights and characterise the vertical distribution of smoke across the Amazon. We provide below a summary of main datasets and tools used in the analysis and compile their main features in Table 3.1.

3.2.1 MINX overview

The MISR Interactive Explorer (MINX) software is an application written in Interactive Data Language (IDL) that is used to analyse the physical properties of smoke plumes and to study plume dynamics (Nelson et al., 2013). MINX can use MODIS thermal anomalies to locate active fires, and MINX then computes the smoke plume
	$\operatorname{Instrument}/$	$Satellite^{a}/$	Level/	Spatial 1	Resolution		
Parameter	Product	Model	Version	Horizontal	Vertical	Temporal Res.	Main reference
Smoke Height	MISR	Terra	Level 1 & 2	$275-500 \mathrm{~m}$	1.1 km	variable	Diner et al.
Aerosol Extinc- tion	CALIOP	CALIPSO	Level 2/v4	30-60 m	333 m	variable	(1998) Winker et al.
Active Fires	MOD14/MYD14	Terra & Aqua	v6	1 km x 1 km		daily	Giglio et al.
Land Cover	MOD12Q1	Terra & Aqua	v5.1	$500 \text{ m} \ge 500 \text{ m}$		annual	(2006) Friedl et al.
PBL	MERRA-2	GEOS-5 DAS	v5.12.4	0.625°x 0.5°	42	hourly	(2010) Bosilovich
Atm. Stab.	MERRA-2	GEOS-5 DAS	v5.12.4	0.625 x 0.5 °	42	6-hourly	et al. (2015) Bosilovich
Drought (DSI)	MOD16/MOD13	Terra & Aqua		0.05°x 0.05°		8-day	et al. (2015) Mu et al. (2013)

Table 3.1: Summary of main features for instruments and products used in the study.

or cloud heights from MISR stereo imagery. MINX also collects particle property results from the MISR Standard aerosol retrieval algorithm (Martonchik et al., 2009). MODIS and MISR are both aboard the NASA Terra satellite, which crosses the equator in the descending node at around 10:30 a.m. local time. These instruments allow temporally and spatially coincident detection of active fires and their associated smoke plumes (Kahn et al., 2008).

MODIS has a cross-track swath of 2330 km that provides global coverage every one to two days. The instrument has 36 spectral channels with wavelengths between 0.4 μ m and 14.2 μ m, and detects thermal anomalies at 1 km spatial resolution (at nadir), under cloud-free conditions. MODIS reports fire radiate power based on a detection algorithm that uses brightness temperature differences in the 4 μ m and the 11 μ m channels (Giglio et al., 2003); this FRP parameter is used as an indicator of fire location and qualitative intensity. We use MODIS Collection 6 (Table 3.1). We note that MINX provides FRP values in MW, although they are actually in MW per 1-km pixel, which corresponds to W/m², except toward the edges of the swath.

MISR has nine push-broom cameras placed at viewing angles spanning -70.5 to 70.5 relative to nadir in the satellite along-track direction (Diner et al., 1998). The cameras each provide imagery in four spectral bands (446, 558, 672, and 867 nm), which makes it possible to distinguish aerosol types qualitatively (Kahn and Gaitley, 2015) and surface structure from the change in reflectance with view angle. This passive stereoscopic imagery method produces cloud and aerosol plume heights, along with cloud-tracked winds aloft. MISR has a swath of 380 km common to all cameras, so global coverage is obtained every nine days at the Equator and every two days at the poles (Diner et al., 1998). The MISR Standard stereo-height product provides vertical resolution of 275–500 meters and horizontal resolution of 1.1 km (Moroney et al., 2002a, Muller et al., 2002).

MINX has a graphical user interface that displays the nine MISR multi-angle images. They can be visualised one by one or as an animated loop, providing a 3-D view of the plume that can help in assessing its structure and dynamical behaviour. In addition, MODIS thermal anomalies can be superimposed, which helps identify the locations of smoke sources from active fires. A user needs to digitise the boundaries of the plume, starting at the source point, and to indicate the direction of smoke transport. The MINX stereoscopic algorithm also calculates wind speed from the displacement of plume contrast elements, which is used subsequently to compute wind-corrected heights, accounting for displacement due to the proper motion of the plume elements between camera views. As with the MISR Standard stereo-height product, MINX automatically retrieves smoke plume heights and wind speed at a horizontal resolution of 1.1 km and vertical resolution of 250–500 m, but with greater accuracy for the plume itself, due to the user inputs (Nelson et al., 2013). MINX plume heights are reported above the geoid, which correspond to the level of maximum spatial contrast in the multi-angle imagery, typically near the plume top, but actually offering a distribution of heights in most cases, because aerosol plumes are rarely uniform (Flower and Kahn, 2017). Additionally, MINX provides local terrain height from a digital elevation map (DEM) product. Here we report heights above the terrain, by taking account of the DEM values. Further information from the MISR Standard Aerosol product about aerosol amount and type is collected and reported, along with FRP from MODIS (Nelson et al., 2013). MINX has been successfully used to investigate fire smoke plume heights over many regions across the world (eg, Kahn et al., 2008, Val Martin et al., 2010, Tosca et al., 2011, Jian and Fu, 2014).

There are several limitations to the MISR-MINX approach that must be considered when studying smoke plume heights. For example, MISR obtains global coverage only about once per week, and the Terra overpass time in late morning does not coincide with the typical, late-afternoon peak of fire intensity. MODIS does not observe FRP under cloud and dense smoke, and the MINX operator must decide whether to include any pyro-cumulus clouds in the plume-height retrieval. These are the key limitations; they and others are discussed further in the literature (eg, Kahn et al., 2007, Val Martin et al., 2010, Nelson et al., 2013). In addition, three MINX versions were used to generate the data in this study, which might introduce an additional bias. MINXv2 and v3 included only MISR red-band plume height retrievals, whereas MINXv4 considers both red and blue-band images. Over land, digitalisation with the blue band usually provides higher quality retrievals, especially for optically thin plumes over bright surfaces. In contrast, red-band provides higher vertical resolution over dark surfaces and sometimes performs better for optically dense smoke layers (Nelson et al., 2013). We take these limitations into account throughout our analysis.

3.2.2 MINX smoke plume database

We limited our study to the burning season (July–November) for the period of 2005–2012. Using MINX, we developed a climatology of plume heights across the Amazon, consisting of 10,858 smoke plumes in the region ($25^{\circ}S-5^{\circ}N$ latitude and $80^{\circ}W-40^{\circ}W$ longitude). Over this domain, the NASA Terra satellite overpass is every 4–8 days at 10:00–11:00 local time. Table 3.2 summarises the number of smoke plumes in each year and the digitising source. The climatology includes a combination of smoke plumes extracted from different projects and created with different versions of MINX (v2–4): plumes for August–September in years 2006 and 2007 are from the MISR Plume Height Project (Nelson et al., 2013); plumes in year 2008 are from the global digitalisation effort made for the AeroCom project (MPHP2 and Val Martin et al. (2018a)); and the five remaining years and additional months are digitised as a part of the current project.

MINX computes several plume heights that describe the altitude that smoke reaches in the atmosphere. In this work, we use the best estimate maximum and median smoke plume heights, which represent the distribution of stereo heights, obtained at the level of maximum spatial contrast over the plume area (Nelson et al., 2013). In addition, as in previous studies, we remove smoke plumes with poorquality retrieval flags. This screening leaves a total of 5393 plumes, about 56% of

		Number of Plum	es^a		
Year	Total	Blue Band	Red Band	MINX	Reference
				version	
2005	927	122	805	v3/v4	This study
2006	513	501	12	v2/v4	$MPHP^{b}/This$
					study
2007	858	670	188	v2/v4	$MPHP^{b}/This$
				,	study
2008	889	889	0	v3.1	$MPHP2^{c}$
2009	150	55	95	v3/v4	This study
2010	1373	0	1373	v3	This study
2011	320	320	0	v4	This study
2012	363	30	333	v3/v4	This study
2005-2012	5393	2587	2806		

Table 3.2: Summary of MISR smoke plumes over the Amazon domain (2005–2012)

^aTotal number of plumes, and number of plumes digitised with blue/red band retrievals

^bMISR Plume Height Project; data from https://misr.jpl.nasa.gov/getData/accessData/ MisrMinxPlumes/

^cMISR Plume Height Project2; data from https://misr.jpl.nasa.gov/getData/accessData/ MisrMinxPlumes2/

the original database, with 77% and 23% plumes digitised in the red and blue bands, respectively. Our final dataset includes plumes digitised in years with intense fire activity associated with severe drought conditions (e.g., 2005, 2007 and 2010) (Chen et al., 2011), in years with low fire intensity and considerable precipitation (2009 and 2011) (Marengo et al., 2013) and in one year when land-management policy measures limited deforestation (2006) (Nepstad et al., 2006). Thus, our climatology is intended to capture smoke plumes variability under diverse conditions.

As mentioned in section 3.2.1, the MISR colour band image used by the MINX algorithm to compute smoke plume heights influences the quality of the plume height and wind speed retrievals. A large majority of the fires detected across our domain have optically thin smoke plumes. Thus, blue band plume retrievals are more successful, with about 60% of the smoke plumes receiving good or fair quality flags, compared to 36% for the red band retrievals. In our dataset overall, most of the plumes were digitised from red band images, as it was the default option for MINX v2–3. However, whenever both band retrievals are available for a plume, blue band is preferred in this study. The choice of the band colour for the retrievals does not affect significantly the results presented here, as the difference in heights for smoke

plumes digitised with both bands is negligible (~ 60 m), lower than the ± 250 m MINX uncertainty.

3.2.3 Land cover unit data

We use the MODIS Level 3 land cover product MCD12Q1 (Friedl et al., 2010) to determine the type of land cover associated with each of our fire smoke plumes. This product contains 17 International Geosphere-Biosphere Programme (IGBP) land cover classes, at a horizontal resolution of 500 m and annual temporal resolution, from 2001 to present day. It is available from the Land Processes Distributed Active Archive Center¹. We merge land cover classes having similar characteristics into four land types representing the main biomes across the Amazon: tropical forest, savanna, grassland and crops.

3.2.4 Atmospheric conditions

To assess the role of atmospheric conditions on the final elevation of smoke plumes across the Amazon, we analyse data from the second Modern Era Retrospectiveanalysis for Research and Applications (MERRA-2) reanalysis model simulation (Bosilovich et al., 2015). We focus on the height of the planetary boundary layer (PBL) and the atmospheric stability at the location of our fires. As in previous studies (eg, Kahn et al., 2007, Val Martin et al., 2010), we define the atmospheric stability as the vertical gradient of potential temperature. We use data from MERRA-2 at a horizontal resolution of 0.625°longitude by 0.5°latitude, with 42 levels vertical pressure-levels between the surface and 0.01 hPa. MERRA-2 provides hourly PBL height above ground level and potential temperature profiles every 6 hours (0:00, 06:00, 12:00 and 18:00 UTM), so we linearly interpolate these data to the time and location of each fire plume origin.

¹https://lpdaac.usgs.gov/get_data

3.2.5 Drought conditions

To determine the presence and magnitude of droughts over the Amazon during our study period, we use the MODIS Drought Severity Index (DSI). The DSI is a global drought index derived by combining the MODIS16 Evapotranspiration (e.g., Mu et al., 2007) and the MODIS13 vegetation index (NDVI) data products (Huete et al., 2002). DSI provides drought conditions at global scale for all vegetated areas at 8-day and annual temporal resolutions and 0.5 or 0.05°horizontal spatial resolution for 2000–2011 (Mu et al., 2013). In this work, we use the 8-day temporal resolution DSI and interpolate the data to the time and location of our fire smoke plumes. Following Mu et al. (2013), we further define drought conditions as: "Extreme-Severe" (DSI ≤ -1.2), "Mild-Moderate" ($-1.2 \leq DSI < -0.29$), "Normal" (-0.29 > DSI > 0.29) and "Wetter than Normal" (DSI ≥ 0.29).

3.2.6 CALIOP observations

We also use extinction profiles derived from the CALIOP instrument to provide an independent assessment of the vertical smoke distribution across the Amazon. CALIOP is a space-borne two-wavelength polarisation lidar (532 and 1064 nm) that flies aboard the CALIPSO satellite (Winker et al., 2013). CALIPSO was launched in 2006 into a sun-synchronous polar orbit of 705 km altitude as a part of the "A-Train" constellation, with an orbit repeat cycle of 16 days. CALIOP collects backscatter and depolarization data that constrain the vertical structure and some properties of aerosols and clouds around the globe (Vaughan et al., 2004, Liu et al., 2009). In addition, CALIOP provides a characterisation of the aerosol type (i.e. dust, polluted dust, marine, clean continental, pollution and biomass burning) based on externally determined surface type along with measured depolarisation ratios, integrated backscatter altitude and colour ratio (Omar et al., 2009). This aerosoltype classification can be used to indicate the likely sources that contribute to aerosol mass loading at specific locations and times where the instrument has coverage. We use CALIOP Level 2 version 4 day and night data (CAL_LID_L2_05kmAPro-Standard-V4–10) over the Amazon for the July to November burning season, from 2006–2012. In this work, we filter the data following Ford and Heald (2012). This filtering approach uses cloud-aerosol distinction scores, extinction uncertainty values, atmospheric volume descriptors, extinction quality control flags and total column optical depths, and assumes that extinction observations classified as 'clear air' have zero aerosol extinction (rather than the fill value). CALIOP daytime retrievals can be biased low due to the noise from scattered solar radiation (Winker et al., 2009, Rogers et al., 2011). However, we analyse both day (i.e., early afternoon, $\sim 13:30$ LT equator crossing time) and night profiles to identify any differences in smoke heights, as well as to allow a better comparison with the MISR smoke plumes, which are retrieved during the late morning.

The CALIOP "swath" is ~100 m wide, so sampling is effectively a curtain. To obtain a climatology of CALIOP smoke plumes as in MISR, we developed an approach to identify individual smoke plumes in the CALIOP data. We first grid all CALIOP aerosol extinction profiles classified as smoke (day and night) at a horizontal resolution of $0.5^{\circ}x \ 0.5^{\circ}$ over the Amazon region, and a vertical resolution of 250 m, from the surface to 12 km. We chose this horizontal resolution to optimise computing processing time. Within each grid cell, we then determine the vertical distribution of smoke extinction. We define the maximum smoke plume height in each grid cell as the maximum altitude reached by the extinction classified as smoke. Similar to the MINX definition of median plume height, we consider the median of the CALIOP vertical extinction distribution as the height where most of the smoke is probably concentrated. Smoke does tend to concentrate either in the PBL or in thin layers in the FT (Kahn et al., 2007, Val Martin et al., 2010).

To ensure we do not introduce a bias in the CALIOP plume heights due to the $0.5^{\circ}x \ 0.5^{\circ}$ horizontal resolution, we also retrieved the smoke plumes for the 2017 burning season at a horizontal resolution of $0.1^{\circ}x \ 0.1^{\circ}$, and find no significant differences. For this subset, our $0.5^{\circ}x \ 0.5^{\circ}$ method returns 131 plumes, with an average altitude of 3.65 km for the maximum plume heights, whereas the $0.1^{\circ}x$ 0.1° method returns 149 plumes, with an average altitude of 3.74 km.

To identify CALIOP smoke plumes associated with active fires, we select only those CALIOP-derived grid cells that contain at least two MODIS Collection 6 fire pixels (Giglio et al., 2003), at 80% confidence level or higher, at the time of CALIOP overpass. We also use the mean terrain elevation across each grid cell to reference the maximum and median heights to ground level, as CALIOP provides observations above sea level. We estimate the mean terrain elevation using terrain elevation from the CALIOP digital elevation map. Figure 3.1 shows an example of our approach for the CALIOP observation of September 25th, 2010 at 06:25 UTC. For this example, we identify a CALIOP smoke plume with 1.7 km median and 4.5 km maximum height above ground level. A total of 2460 plumes are characterised with our approach over the Amazon for the months of July to November, from 2006– 2012; about 65% of these plumes are linked to actives fires with some confidence (i.e., having a clear connection to a MODIS fire pixel), and we only consider those in our analysis, a total of 1600 plumes.

Previous studies used other CALIOP products to determine the vertical distribution of smoke plumes. The Level 2 Aerosol Layer product is commonly used to analyse smoke plume heights from CALIOP, as it reports the top and base heights of aerosol layers. Tosca et al. (2011) used their smoke layer top altitudes and extinction coefficient profiles over Borneo for September–October 2006. Using the CALIOP Level 1 attenuated backscatter profiles at 532 nm, Amiridis et al. (2010) estimated smoke injection heights from agricultural fires over Europe. They selected only those profiles of constant attenuated backscatter coefficient with height, without strong convection, and that were collocated with MODIS active fire pixels from the Aqua satellite. Recently, Huang et al. (2015) used six years of the CALIOP Level 2 vertical feature mask (VFM) data and aerosol layer products over six regions to investigate the Most Probable Height (MPH) of dust and smoke layers. They used two approaches to obtain MPH: one based on the probability distribu-



Figure 3.1: Example of the approach followed for the CALIOP smoke plume characterisation. The map shows estimated smoke plume median heights (gridded at 0.5x0.5 horizontal resolution) for September 25th, 2010 at 06:25 UTC. MODIS active fire pixels associated with the CALIOP smoke plumes are represented with open circles. The insert displays the vertical distribution of aerosol extinction for a specific smoke plume in the map, with extinction values coloured by classified aerosol types. Dashed black line represents the averaged extinction profile for the aerosols classified as smoke (pink dots). In this profile, the CALIOP smoke plume has a median height of $\sim 2 \text{ km}$ (green colour in the smoke plume median height scale) and a maximum height of 4.5 km above the terrain.

tion of the vertical profiles of Occurrence Frequency (OF) (i.e., ratio of number of samples classified as dust or smoke by the VFM to the total samples per grid) and the other as the probability distribution of the aerosol optical depth (AOD) vertical profiles. So MPH_OF and MPH_AOD correspond to the altitude with the largest OF and mid-visible AOD for a certain type of aerosol. Our definition of CALIOP median plume height is most similar to their MPH_AOD. However, Huang et al.

(2015) analysed vertical profiles over large-scale regions (e.g., the entire Amazon or Sahara), whereas in the current work, we analysed and then aggregated the heights for individual smoke plumes.

Our initial objective was to compare the CALIOP with the MISR plumes to assess the diurnal smoke evolution, as CALIOP has a later sampling time than MISR over the Amazon (14:00–15:00 LT versus 10:00–11:00 LT). However, despite our effort to develop a comprehensive CALIOP climatology, none of the CALIOP plumes coincide with the MISR plumes. As previous studies discuss (eg, Kahn et al., 2008, Tosca et al., 2011), CALIOP and MISR, in addition to having different sampling times, also have different swath widths (380 km versus 70 m). These differences make it difficult to observe the same fire on the same day, but they make CALIOP and MISR observations complementary: MISR provides late-morning near-source constraints of aerosol plume vertical distribution, whereas CALIOP in general offers more regional constraints, later in the day (Kahn et al., 2008). Some differences between the products are thus expected.

3.3 Results and discussion

3.3.1 Smoke plume height observations

Figure 3.2 maps the biomes of the Amazon region for which the MISR plume climatology was developed. Figure 3.3 presents the time series of the smoke plume heights for the biomass burning seasons (July–November) during the 2005–2012 study years. We also include a statistical summary of the number of plumes within the time series by year, month, biome and drought conditions in Figure A1. The largest number of plumes is recorded in 2010, with about 25% of the total plumes in the database, whereas the smallest is in 2009 (3%). These two years are the driest and the wettest in the climatology, respectively. Most of the plumes were observed in August and September (85%), at the peak of the burning season in most vegetated locations, in the dominant biomes of savanna (48%) and tropical forest (46%), and during dry conditions (76%). We find important interannual variability in the type of fires, with fires over tropical forest dominant in 2005 (65%) and 2010 (47%), two of the three drought years in our database as shown in Section 3.3.4 below, and the majority of fires in savanna (54–65%) for the rest of the years. We note that a large fraction of the plumes were observed in 2008 (17%) even though it was not a drought year. The majority of plumes in the 2008 record are digitised with blue band retrievals (Table 3.2), which produce higher quality results in many situations, especially for optically thin plumes over land surfaces.



Figure 3.2: Locations of the MISR plumes analysed (black dots) over the four main biomes considered in the study. The black square represents the Amazon domain.

Throughout the study period, we find significant variability in smoke plume height, with altitudes ranging from a few meters (essentially near-surface) to 5 km, depending on the biome (Figure 3.3). Smoke plumes over cropland fires are scarce compared to the other fire types, as these fires are small and tend to be under-



Figure 3.3: Time series of the 2005–2012 MISR Amazon smoke-plume-height climatology, covering the July-November burning season for each year. Each blue dot represents the maximum smoke height above ground level (agl) for one plume.

detected by MISR (Nelson et al., 2013). We summarise in Table 3.3 the statistical parameters of the smoke plumes for all observations except the cropland cases. Over the Amazon, the vertical distribution of smoke varies by biome. Statistically, the highest smoke altitudes averaged by biome are detected over grasslands, with median and maximum heights of 794 m and 1120 m, respectively, whereas the lowest heights are detected over tropical forest (601 and 845 m, respectively). In all the biomes, more than 85% of the smoke is located at altitudes below 2 km (Figure A2).

Table 3.3:	Statistical	summary for	main sı	moke plı	ume para	meters and	l atmospl	neric
$conditions^a$.								

	Tropical Forest	Savanna	Grassland
Median Height (m)	601 ± 339	743 ± 422	794 ± 471
Max Height (m)	845 ± 499	1040 ± 585	1120 ± 653
MODIS FRP (MW)	209 ± 537	360 ± 658	421 ± 614
AOD (unitless)	0.51 ± 0.34	0.33 ± 0.28	0.35 ± 0.29
Atm Stab (K/km)	4.21 ± 2.97	3.16 ± 3.16	2.52 ± 2.50
BL Height (m)	1270 ± 514	1490 ± 507	1620 ± 530
Plumes in FT $(\%)^b$	3 - 15	4 - 17	5 - 19
Number	1744	2084	166

^aReported the average \pm SD and number of observations

 b Reported range from more and less conservative definition of plume in the FT (see text for explanation).

Similar altitudes and distributions have been found across comparable fires in

other parts of the world. For example, altitudes between of 700–750 m were detected over the tropical forest in central America and Indonesia (Val Martin et al., 2010, Tosca et al., 2011). In contrast, smoke plume heights over the Amazon are substantially lower than smoke plumes observed over the boreal biomes (960–1040 m) (Kahn et al., 2008, Val Martin et al., 2010). There are several factors that influence smoke altitudes and contribute to the differences between biomes, such as fire intensity, availability of fuel, combustion efficiency, atmospheric stability, and entrainment (e.g., Lavoué et al., 2000, Trentmann et al., 2006, Luderer et al., 2006, Kahn et al., 2007, 2008, Val Martin et al., 2012). We assess some of these factors for our Amazon dataset next.

3.3.2 Effect of atmospheric and fire conditions on smoke plumes

We explore the relationship between smoke plume height, fire characteristics (i.e., MODIS FRP and AOD) and atmospheric conditions derived in the vicinity of the fires throughout the burning season, across the major biomes in the Amazon except cropland. For atmospheric conditions, we focus both on how smoke plume height relates to boundary layer height and on the effect of atmospheric stability on plume rise. We consider atmospheric stability conditions above our fires as the average of the atmospheric stability over the atmospheric column (K/km; Section 3.2.4) from the surface, at the origin of the fire, to the maximum altitude that smoke reached in the atmosphere. We add a buffer of 10% to the maximum altitude to account for any potential influence that the atmosphere above the plume might have over the column. We include in Table 3.3 a summary of these main parameters.

Consistent with previous studies (e.g., Val Martin et al., 2010, 2012, Sofiev et al., 2009, Amiridis et al., 2010), we find that the highest-altitude smoke plumes tend to be associated with highest MODIS FRP values, though there is significant variability in the relationship in all the biomes ($r^2=0.2$; Figure A3). Smoke plumes detected

over tropical forest fires have the lowest FRP (209 MW) and largest AOD values (0.51) (Table 3.3). The other two main biomes (savanna and grassland) have FRP and AOD values similar to each other (360–421 MW and 0.33–0.35, respectively). Tropical forest has deeper root systems, which allows fires to access deeper soil layers (Nepstad et al., 2008) that can maintain higher moisture content and lower oxygen availability than other biomes, such as grasslands. High fuel moisture content and low oxygen availability favour smouldering rather than flaming fires, which in turn tends to produce greater smoke emission but lower radiant emissivity (Kauffman et al., 1995). Therefore, the low FRP and high AOD in tropical forest fires are consistent with these conditions, in which smouldering fires predominate, whereas high FRP and low AOD are typical with dryer, less dense fuels, eg, savanna and grassland, that tend to produce flaming fires (Giglio et al., 2006). In addition, high smoke opacity and tree canopy obscuring the fire-emitted 4-micron radiance as viewed by MODIS, as well as low radiant emissivity, rather than just low radiative total fire intensity, probably contribute to these differences (Kahn et al., 2008).

The atmospheric stability structure affects the vertical motion of smoke and is a key factor in plume rise, either enhancing or suppressing the lifting. Some studies have shown the important role that atmospheric stability plays in plume rise (e.g., Kahn et al., 2007, 2008, Val Martin et al., 2010, Amiridis et al., 2010), and the quantitative representation of this factor still remains an open question in plumerise model parametrisations. For instance, Val Martin et al. (2012) showed that, in North America, fires that inject smoke to high altitudes tend to be associated with higher FRP and weaker atmospheric stability conditions than those that inject smoke at low altitudes, in which smoke tends to be trapped within the boundary layer. Similar results were found for agricultural fires over eastern Europe (Amiridis et al., 2010).

To analyse the influence of atmospheric stability over Amazon fires qualitatively, we divide our plume dataset into two groups that we define as having weak and strong atmospheric stability conditions based on MERRA-2 reanalysis. Over the



Figure 3.4: Vertical distribution of MISR stereo-height retrievals for all the plumes analysed, under strong (blue) and weak (pink) atmospheric stability conditions.

Amazon, and at the locations and times studied, atmospheric stability ranges from -3 to 23 K/km. We designate atmospheric stability < 2 K/km as 'weak', and atmospheric stability > 4 K/km as 'strong'. Each group contains about 30% of plumes in the database. Figure 3.4 shows the vertical distribution of smoke stereo-height retrievals for the plumes classified under weak and strong atmospheric stability conditions. Our comparison supports previous observations that plumes under weak atmospheric conditions tend to inject smoke to higher altitudes than those encountering strong stability, with maximum plume heights of 1150 m and 654 m, respectively. A similar pattern is found for the median plume heights (821 and 482 m, respectively). Weak atmospheric stability conditions are also associated with deeper PBLs (~1500 m) than strong stability conditions (~1200 m) (not shown).

Atmospheric conditions also correlate with biome type. We find that tropical forest fires tend to be associated with more stable atmospheric conditions than grassland fires (4.2 versus 2.5 K/km). Shallower PBLs are also observed over tropical forest (1270 m) compared to grassland (1620 m). Tropical forests typically have higher relative humidity conditions and more constant temperatures than grasslands, which favours more stable conditions and lower PBL heights (Fisch et al., 2004). We note that our dataset was all acquired at Terra overpass time, which occurs

between about 10–11 am LT. This might produce a bias toward the more stable atmospheric conditions that occur preferentially during the morning; later in the afternoon convection tends to become more important (Itterly et al., 2016).

3.3.3 Seasonality of smoke plumes heights

Figure 3.5 shows the seasonal cycle of maximum plume height with FRP, AOD, and atmospheric conditions over the major Amazon biomes. We further disaggregate these observations by biome, season and dry/wet years in Table A1. For these biomes, we find minimum plume heights of 600–750 m in July and maximum plume heights of 900–1400 m in October and November. Similarly, over tropical forest and grassland, MODIS FRP values follow the plume-height patterns, with maximum values toward the end of the burning season (180–200 MW), compared to the early season (90 MW). For savanna fires, MODIS FRP remains mostly constant throughout the season (~150-200 MW). Savannas are known to be fire-adapted, and combustion efficiency typically remains constant throughout the season (Van der Werf et al., 2010). All these patterns are similar in wet and dry years, although larger MODIS FRP values are observed over savanna and grassland fires in dry years (Table A1).

Some previous studies show the seasonal peak in MODIS FRP over the Amazon earlier, in August–September (Tang and Arellano, 2017). However, their work relies on the maximum MODIS FRP detected by the Terra and Aqua satellites (four times/day) over the Amazon, whereas our seasonality shows the monthly median MODIS FRP observed by Terra, collocated with the MISR smoke plume observations (once/day). In addition, the MISR swath is substantially narrower than MODIS (380 versus 2330 km), and many fires detected by MODIS are not observed by MISR. Our seasonality thus captures the fire intensity that drives the smoke plumes detected specifically by MISR, i.e., only at about 10:30 AM local time, and the seasonal differences provide at least some indication of possible bias introduced



Figure 3.5: Seasonal cycle of MISR smoke plume maximum height above the terrain (black circles), MODIS FRP (red diamonds), PBL heights (black triangles), atmospheric stability (blue diamonds) and MISR AOD (green diamonds). Monthly median values are shown for tropical forest, savanna and grassland biomes. Vertical bars indicate the 10th and 90th percentile. Distributions with fewer than 10 observations are omitted and all years are included

by the MISR sampling of fires.

In contrast to the seasonality of plume heights and fire intensity, the peak monthly AOD occurs in September across the major biomes, with median AOD of 0.6 in tropical forest and 0.3 in savanna and grasslands, compared to AOD values of 0.04–0.1 in July and November. Over the Amazon, total AOD correlates well with the number of fires, and both tend to peak during September each year (Mishra et al., 2015). Baars et al. (2012) reported optical depths in the polluted biomass burning season (July–November) six times larger (on average) than in the pristine wet season (December–June), with highest values in September and October, for a site in the central Amazon near Manaus. In our dataset, September, together with August, are the months when the largest number of plumes were detected (Figure A1). However, our monthly statistics might be influenced by the number of observations in each month. For example, the number of fires in August is driven by year 2010, in which an unusually large number of fires were observed, compared to the other August months. In addition, the large monthly median values in November are based on fewest numbers of plumes (Figure A1), although the few fires detected by MISR for those months were large and intense.

Boundary layer heights and atmospheric stability conditions may also vary by biome and throughout the season, influencing plume-rise spatial and temporal distributions. On a seasonal basis, the PBL height does not follow a clear cycle in any of our biomes, but higher PBL heights are observed over grassland fires (Table 3.3) and across all the biomes during dry years (Table A1 and Figure A1). More stable atmospheric conditions are found at the beginning (3.6 K/km in July) compared to the end of the burning season (1.9 K/km in November).

Previous studies have shown that a substantial fraction of smoke is injected above the boundary layer (i.e., into the FT), although this fraction varies depending on biome and fire type. For tropical fires over central America and Indonesia, smoke from about 4-6% of fires is reported to reach the FT (Val Martin et al., 2010, Tosca et al., 2011). This fraction is larger for boreal fires (>16%), where fires are more intense and the PBL is typically lower than in tropical regions (Val Martin et al., 2010, Kahn et al., 2008, Val Martin et al., 2018a). Following these studies, we consider that smoke reaches the FT when the median height of the plume is at least 500 m above the PBL height. This is a conservative definition that takes into account uncertainties in MINX and MERRA (e.g., Kahn et al., 2008, Val Martin et al., 2010, Tosca et al., 2011). Because fires over the Amazon tend to be smaller in size than those in boreal forests, we also consider a less conservative definition. We assume a plume is injected into the FT when the maximum plume height is at least 250 m above the PBL height. We understand that this is an upper limit, but it provides a bracket to our results. We include in Table 3.3 the percentage of the smoke plumes injected into the FT for both definitions, and present in Figure 3.6 the seasonality of these percentages. Our analysis shows that fires at the end of the burning season are more likely to inject smoke in the FT, with 15–40% in November versus 2–10% in July, and 5–22% at the peak of the burning season (August–September). This pattern seems to be related to a combination of more intense fires and less stable atmospheric conditions. We find no influence of the monthly PBL depth variability, although deeper PBL heights are found across the Amazon in drier conditions (i.e., over grassland fires and/or dry years). Interestingly, our analysis also shows a slightly larger percentage of fires injecting smoke into the FT over grassland (5–19%) compared to tropical forest (3–15%). As mentioned above, grassland fires are associated with high PBL heights, but also with large FRP values, suggesting that these fires are energetic enough to produce the buoyancy needed for the smoke to reach the FT.



Figure 3.6: Seasonal variation of Amazon plume injection above the PBL (percent). Bar plots indicate the average of [Median Plume–PBL Height] > 0.5 km (dark grey) and [Maximum Plume–PBL Height] > 0.25 km (light grey) (see text for explanation).

3.3.4 Interannual variability of smoke plumes and drought conditions

We use MODIS DSI to assess the effect of drought conditions on smoke plume rise and the extent that these conditions control the interannual variability of smoke plumes across the region. We present the interannual variability of MISR plume heights, MODIS FRP and MISR AOD in Figure 3.7, and summarise the annual averages of MODIS DSI, atmospheric stability, PBL height and percentage of smoke plumes in the FT in Table 3.4. In addition, we include the annual relationship of MISR plume heights, MODIS FRP and MISR AOD with MODIS DSI, and the percentage of plumes in the FT per drought level in Figure 3.8. In our dataset, 76% of plumes are recorded under extreme-mild drought conditions versus 7% plumes in wet conditions, as discussed in Section 3.3.1. During drought years (2005, 2007 and 2010), smoke plumes register the lowest MODIS DSI annual averages values (-0.89, -0.91 and -1.50, respectively), compared to the other years in the climatology (-0.63–0.18). Note that DSI is higher in wetter years.

Table 3.4: Summary of the main atmospheric parameters calculated at the location of the plumes per year^{*a*}.

		BL height	Atm. Stab	$\%$ in FT^c
Year	Number	(m)	(K/km)	
2005^{b}	927	1370 ± 546	4.32 ± 3.01	3 - 13
2006	513	1210 ± 518	3.50 ± 2.89	6 - 25
2007^{b}	858	1380 ± 539	3.96 ± 3.30	3 - 18
2008	889	1480 ± 558	3.02 ± 2.28	4 - 23
2009	150	1100 ± 377	3.22 ± 2.60	4 - 27
2010^{b}	1373	1550 ± 498	3.69 ± 3.53	$2\!-\!7$
2011	320	1150 ± 296	2.73 ± 2.38	8 - 28
2012	363	1330 ± 453	3.20 ± 3.29	4 - 13

^{*a*}Reported the average \pm SD

^bDrought years

^c Reported as percentage of plumes where [Median Plume-BL Height] > 0.5 km-[Maximum Plume-BL Height] > 0.25 km (see text for explanation).

We find a significant positive relationship between MISR maximum plume heights and MODIS DSI (r=0.7; p<0.01) in tropical forest and savanna fires, with higher maximum plume heights in normal and/or wetter than normal (1000–1100 m) than severe drought conditions (750–900 m) (Figure 3.8). Consistently, on an annual basis, these two biomes show the lowest smoke plume heights during dry years (Figure A1). Smoke plume heights in grassland fires, however, do not show any strong relationship with DSI (r=0.1) or a clear interannual variability driven by droughts (Figure A1). In general, lowest median smoke heights are observed in our dataset



Figure 3.7: Interannual variability of MISR maximum plume heights above the terrain, MODIS FRP and MISR AOD, for the aggregate of tropical forest, savanna and grassland. Bar plots indicate the distribution of the data for each year. The medians (red circles) and the means (black squares) are shown along with the central 67% (box) and the central 90% (thin black whiskers). The number of observations (in black) and the median values (in red) included in each distribution are given at the top of the plot. Drought years are in pink and non-drought years in light blue. The same data, stratified by biome type, are plotted in Figure A4.

during the drought years of 2005 and 2010 (Figure 3.7), which are driven by tropical forest observations as they are the dominant biomes (Figure A1).

The relationship between MODIS FRP and drought levels over the Amazon is not straightforward on an annual basis as we do not observe any clear interannual variability of FRP driven by drought in Figure 3.7. However, our analysis shows some patterns when we subdivide the data by biome (Figures 3.8 and Figure A4). For example, we find a significant positive relationship between MODIS FRP and DSI (r=0.6; p<0.01) in tropical forest, with lower FRP in extreme dry than normal-wet conditions (170 versus ~250 MW; Figure 3.8). Contrariwise, savanna and grassland



Figure 3.8: Relationship between MODIS DSI at the location of the plumes and MISR maximum plume height, MODIS FRP and MISR AOD annually averaged, for tropical forest (green), savanna (blue) and grassland (red). Symbols represent the annual average and bars the standard error of the mean. Regression lines are weighted by the number of plumes in each year; relationships with absolute r < 0.4 are plotted in dashed lines. Also included percentage of smoke plumes in the FT in each biome and by drought condition. Bar plots indicate the average of [Median Plume–PBL Height]> 0.5 km (light colour) and [Maximum Plume–PBL Height]> 0.25 km (dark colour), based on MERRA-2 PBL heights (see text for explanation).

fires have higher FRP in extreme and mild dry than in wet conditions (\sim 500 MW versus 250 MW), although the relationship is weak (r=-0.4; p<0.01). As mentioned above, interpretation of FRP can be complicated by factors such as overlying smoke opacity and fire emissivity (Kahn et al., 2008).

The relationship between smoke plume height, FRP and drought conditions over the Amazon is somewhat complex. Drought conditions over the Amazon increase fuel flammability and the number of fires, but not necessarily increase smoke elevation. Drought also decreases fuel load, i.e., fuel available to burn, especially over grassland. Tang and Arellano (2017) reported that drought in the Amazon favours understory fires for tropical forest, which are dominated by smouldering combustion and are linked to low altitude smoke plumes. In addition, spatial changes in drought location can influence the type of biome affected and hence the type of fire regime in a given year. For example, drought in 2005 was located at the northeastern and central regions, and the large majority of the plumes recorded by MISR (65%; Figure A1) were from tropical forest fires, i.e., related to smouldering and fires that inject smoke to lower altitudes. In 2007, drought shifted to the southeastern region, and the majority of the plumes (60%; Figure A1) were from savanna and grassland fires associated with more flaming burning conditions, i.e., higher FRP and smoke plume altitudes. Our analysis supports this observation. In 2005, a drought year, smallest MODIS FRP (150 MW) and lowest smoke plume heights (750 m) were recorded over tropical forest (Figure 3.8), whereas in 2007, another drought year, larger FRP (500 and 750 MW), associated with higher smoke plume heights (1100 and 1300 m), were recorded over savanna and grassland fires, respectively.

In addition to the influence of drought in controlling the type of fires, drought can also affect atmospheric conditions. We find that during drought years, PBL heights tend to be about 200 m deeper than in wet years (Table 3.4). However, on an annual basis, atmospheric stability does not vary significantly, with values of $\sim 3-$ 4 K/km, across the Amazon for the averaged biomass burning season (Table 3.4). We also observe that a lower percentage of fires inject smoke plumes into the FT in drought compared to non-drought years (2–18% versus 4–28%; Table 3.4). On a biome basis, tropical forest fires inject a larger percentage of smoke plumes into the FT in wet than extreme-dry conditions (27 versus 12%, Figure 3.8), and shallower PBL heights may partially explain the larger percentage of MISR plumes detected in the FT during non-drought years. Contrariwise, grassland fires, although with fewer observations, inject more smoke plumes into the FT during extreme dry than wet conditions (25% versus 13%, Figure 3.8). These fires are associated with high FRP values in dry conditions and this extra fire energy may be enough to produce the buoyancy needed to lift smoke directly into the FT, regardless of the PBL height. Note that in Figure 3.8 (right bottom), we present the data only subdivided by MODIS DSI and biome, regardless of the year, as in the rest of the panels in Figure 3.8.

Consistent with previous studies that have shown significant positive relationships between drought conditions and aerosol loading (e.g., Reddington et al., 2015, Tang and Arellano, 2017), we find a significant relationship between MISR AOD and MODIS DSI on an annual basis in tropical forest and savanna fires (r=-0.7and p<0.01; Figure 3.8). Years with drier conditions have almost a factor of three greater AOD compared with years with wet conditions. Larger aerosol loading in drought periods is likely due to increases in the number and size of fires (e.g., Aragao et al., 2014) and subsequent increases in aerosol emissions. In addition, MISR AOD shows significant interannual variability, with the largest AOD values recorded in 2005, 2007 and 2010 (0.4–0.6; Figure 3.7), and in particular over tropical forest fires (0.6, Figure A4). Our results suggest that fires during drought periods might significantly degrade regional air quality, as they are associated with low smoke altitude and high aerosol loading.

3.3.5 CALIOP smoke plume observations

To further investigate smoke rise over the Amazon, we develop a climatology of smoke plume heights using CALIOP extinction profiles (section 3.2.6). We identify a total of 1600 CALIOP smoke plumes linked to active fires from July–November, 2006–2012 (Figure A5). Although the CALIOP climatology is 1/3 in size of the MISR climatology, these datasets agree well with respect to the temporal and spatial distributions. Similar to MISR, the largest number of plumes corresponds to years 2007 and 2010 (22 and 29%), whereas the lowest records are in 2009 and 2011 (4 and 7%). Most of the CALIOP plumes are also recorded at the peak of the biomass burning season (September; 51%) and over savanna and tropical forest (37 and 57%, respectively) compared to grassland.

Figure 3.9 displays the time series of derived median and maximum heights, for day and night-time observations. We include both daytime and night-time CALIOP observations to assess any day-night differences in smoke plume rise. Similar to the MISR climatology, we find large variability in the CALIOP smoke plume heights; the median heights range from 0.8-4.4 km (daytime) and 1.1-4.5 km (night-time). Maximum smoke plume heights are obviously higher, typically spanning 1.8–5 km (daytime) and 2.4–5.8 km (night-time). About 18 maximum plume height observations fell above 6 km (shown saturated at 6 km in Figure 3.9). Here we examine the vertical distribution of aerosol plumes individually. Ten cases show high altitude smoke (> 6 km) in a layer that extends through the column to near-surface (Figure A6), right panel), implying that smoke from the active fire below was lifted by fire-induced buoyancy, atmospheric processes, and/or both. The remaining cases show that high-altitude smoke was disconnected from the surface smoke layer (Figure A6, left panel), and we suggest that this smoke could be residual smoke from older fires, smoke transported from the source and concentrated in an elevated layer, aerosol that was wrongly classified as smoke by the CALIOP algorithm, and/or the result of CALIOP not being able to detect lower-level aerosol due to thick smoke aloft or the presence of clouds in the column. We include these observations in our



Figure 3.9: Time series of the CALIOP smoke plumes (2006–2012) for daytime and nighttime observations. Each dot represents the maximum (blue) and median (black) smoke plume height above the terrain. Eighteen points for which the CALIOP height exceeds 6 km are plotted at the top of the charts.

analysis, but note that they represent only 1% of the total observations within the climatology and do not significantly impact the overall statistics shown here.

Figure 3.10 summarises the median and maximum heights for the CALIOP smoke plumes per biome, season and wet/dry years. Night-time plume heights are on average ~ 250 m higher than daytime plume heights (Figure 3.9). Differences between day and night-time CALIOP observations have been attributed in the past to a low bias in the daytime retrievals due to noise from scattered solar radiation (e.g., Winker et al., 2009, Huang et al., 2015). Therefore, our observed difference in day and night-time CALIOP plume heights might result from differences in data quality rather than reflecting smoke diurnal variability. We combine day and nighttime CALIOP observations in Figure 3.10 and include the MISR plume heights for comparison. Average CALIOP median plume heights range from 2.1 km (tropical forest and savanna) to 2.3 km (grassland). Maximum plume heights are similar across all biomes (~ 3.2 km). Similar to MISR, CALIOP detects higher smoke plumes



Figure 3.10: Average CALIOP and MISR plume heights per biome, time of the season and dry/wet years. The burning season is divided into early (July–August–September) and later (October–November) periods, and dry years (2007, 2010) and wet years (2006, 2008, 2009, 2011). Bars represent MISR plume heights (grey), and combined day and night-time CALIOP plume heights (red).

during the late burning season (2.1 and 3.3 km, for the median and maximum plume heights, respectively) than the early season (1.9 and 3.0 km). In contrast, CALIOP observes smoke at higher altitudes during dry (2.2 and 3.4 km) than wet years (2.0 and 3.2 km). In contrast, CALIOP observes smoke at higher altitudes during dry (2.2 and 3.4 km) than wet years (2.0 and 3.2 km). As discussed above, for the time and location of the MISR observations, a deeper PBL is observed in dry compared to wet years. Likewise, PBL heights at the CALIOP smoke plumes are 2.4 and 2.6 km in wet and dry years, respectively, and thus a deeper PBL during drought conditions explain the higher altitudes observed by CALIOP under drier conditions.

Smoke plume height values over the Amazon similar to ours were reported in other studies for CALIOP (Huang et al., 2015) and surface-based lidar measurements (Baars et al., 2012). Using the CALIOP vertical feature mask and AOD profiles, Huang et al. (2015) reported an average for the most probable smoke height of 1.6-2.5 km for September fires. Their definition is comparable to our CALIOP median plume height, which produced a value of 2.3 ± 0.7 km for the September months. Over Manaus in 2008, Baars et al. (2012) reported biomass burning layers at 3–5 km elevation, with most of the smoke trapped below 2 km. Other CALIOP smoke plume heights have been reported over eastern Europe (1.7–6 km) and several

regions and biomes across Asia (0.8–5.3 km)(Amiridis et al., 2010, Labonne et al., 2007, Tosca et al., 2011, Vadrevu et al., 2015).

In our study, CALIOP observes smoke at systematically higher altitudes than MISR, with median plume heights up to 1.4 km higher (2.2 km for the maximum plume heights). However, CALIOP still shows that the majority of the smoke is located at altitudes below 2.5 km above ground, consistent with previous observations from lidar measurements (Baars et al., 2012). Differences between MISR and CALIOP smoke plume heights are consistent with deeper PBL heights at the time of the CALIOP observation, as PBL is expected to grow further later in the day, and fires might also increase in intensity. We find that PBL height at the location/time of the CALIOP daytime smoke plumes is on average about 1.4 km higher than for MISR smoke plumes, specifically 2.7 km for CALIOP and 1.3 km for MISR.

Tosca et al. (2011) found similar differences between CALIOP and MISR (1– 2.8 km) in peatland fires over southeastern Asia. In addition, CALIOP height retrievals are more sensitive to thin aerosol layers than MISR stereo analysis, so CALIOP is more likely to detect low-density smoke at plume-top (Kahn et al., 2008); this would include smoke that might have been lifted later in the day by convection, air mass advection or fire buoyancy (Kahn et al., 2008, Tosca et al., 2011). Although we only select CALIOP plumes that are directly linked to active fires with some confidence, fires can burn for several days (and even weeks); in particular, deforestation fires can leave residual smoke layers over the region for many days or even weeks. As such, our CALIOP plume heights may include lowdensity smoke at higher altitudes, possibly from old fires.

Some previous studies with MISR smoke plume height have also analysed the altitude of 'smoke clouds', that is, dispersed smoke not easily associated with a particular fire (Val Martin et al., 2010, Tosca et al., 2011). Smoke clouds tend to occur at higher altitudes than smoke plumes; they typically represent fire plumes at a later stage of evolution. Over Borneo peatland fires, Tosca et al. (2011) show that MISR smoke clouds and CALIOP smoke plumes had similar altitudes during their

period of study. The analysis of smoke clouds over the Amazon may support the expectation that plume heights tend to grow even larger than observed by MISR later in the afternoon. In addition, transported smoke is more likely to have stayed aloft longer than near-source smoke, and would therefore have more opportunity to mix upward.

3.4 Conclusions

A climatology of smoke plumes from MISR and CALIOP observations is used to characterise the magnitude and variability of smoke altitude across the Amazon during eight biomass burning seasons. Biome type, fire and smoke properties (FRP and AOD), atmospheric conditions (PBL height and atmospheric stability) and regional drought state are included in the analysis, to explore the degree to which each contributes to the observed variability.

Analysis of the smoke plume climatology shows large differences in smoke-plume elevation over the main biomes in the Amazon, with heights ranging a few hundred meters to 5.2 km above ground level. Smoke from plumes observed by MISR (10:00-11:00 LT) is mainly concentrated at altitudes below 1.5 km. As expected, smoke plume elevations are higher in our CALIOP climatology, ranging from 0.8 to 6 km during daytime (14:00-15:00 LT), although the majority are concentrated below 2.5 km. We find that CALIOP smoke plume heights are about 1.4–2.2 km higher than MISR smoke plumes, due to a deeper PBL later in the day, possibly more energetic afternoon fires and CALIOP's greater sensitivity to very thin aerosol layers (Kahn et al., 2008, Flower and Kahn, 2017). Thus, our CALIOP plume climatology includes fresh smoke from active fires and low-density smoke at higher altitudes, some of which might be from old fires. Our results show that over the Amazon, and similar to other fire regions studied previously, on average, smoke plume heights tend to increase later in the afternoon due to greater near-surface convection, greater fire intensity, and possibly self-lofting. Direct injection of smoke to altitudes higher than 6 km (middle to upper troposphere) did not seem to be significant over the Amazon during our study period.

For our main biomes in the Amazon, smoke plume heights are substantially lower over moist tropical forest fires (0.8 km, maximum plume height definition) than grassland fires (1.1 km), although grassland smoke fire plumes represent a small fraction (4%) of cases in the climatology. The MISR and CALIOP Amazon plume climatologies show a well-defined plume height seasonal cycle in the main biomes, with larger heights toward the end of the burning season. Using MODIS FRP and MERRA-2-estimated atmospheric stability conditions, we determine that higher smoke-plume elevations in October–November are the result of the combination of more intense fires and a less stable atmosphere. Less than 5% of the fires inject smoke into the FT (i.e., Median Plume–PBL height > 500 m) using a conservative criterion, although an additional 15–19% of the fires may inject some smoke based on a looser criterion (i.e., Maximum Plume–PBL height > 250 m). This fraction increases throughout the burning season, with about 15–40% of the fires injecting smoke above the FT in November.

Previous studies have shown a direct connection between drought, large-scale climate processes (e.g., ENSO) and the number of fire occurrences (e.g., Alencar et al., 2006, Inness et al., 2015). We find a negative relationship between MISR plume heights and drought conditions in tropical forest fires, as wet years show smoke plume altitudes 300 m higher than dry years. Tang and Arellano (2017) reported that drought conditions over the Amazon favour understory fires, for which smouldering combustion dominates, favouring lower smoke injection heights. In addition to low-altitude smoke, we find that drought conditions are also related to deeper PBL heights, which can reduce the frequency with which smoke is able to reach the FT.

A relationship between fire intensity (as approximated by FRP) and drought conditions is not clear in our study. We detect the highest FRP values in grassland fires during dry periods, and the lowest FRP values for tropical forest fires under similar dry conditions, but without a significant relationship between FRP and DSI, nor any interannual variability of FRP driven by droughts. This lack of relationship may be due to the different locations of drought in different years, the type of fires recorded by MISR in a given year, and/or the low performance of MODIS FRP under dense smoke conditions.

Consistent with previous observations, we find larger MISR AOD during drought compared to non-drought periods. Our analysis confirms the important effect that biomass burning has on smoke aerosol loading over the region, from the surface to the lower free troposphere. Strong land management policies to control fires over the Amazon may become crucial as increases in drought frequency are projected in a future climate (Malhi et al., 2008); this would have important consequences for fire activity and thus air quality.

A variety of smoke injection height schemes are used to represent fire emissions over the Amazon, from fire emissions injected below 3 km (Reddington et al., 2016) or into the model-defined PBL (Zhu et al., 2018) to complex plume rise models, in which a significant fraction of emissions are in some conditions injected above 6 km (Freitas et al., 2007). Recent efforts have shown the value of using MISR-derived smoke plume heights to initialise model fire emission injection (Vernon et al., 2018, Zhu et al., 2018). Over the Amazon, Zhu et al. (2018) show that a new injection scheme based on MISR plume-height observations, which included vertical smoke profiles used in this study (Val Martin et al., 2018a), provide a better representation of CO observations over the region. With a very narrow swath but sensitivity to sub-visible aerosol, CALIOP tends to sample aerosol layers downwind, providing information complementary to the near-source mapping offered by MISR (Kahn et al., 2008). Thus, observations from both CALIOP and MISR provide a way to study smoke plume heights across the Amazon during the biomass burning season. Ultimately, this information will help improve the representation of biomass burning emissions in Earth system atmospheric models, and should aid our understanding of the feedbacks between drought, terrestrial ecosystems and atmospheric composition over the region.

A next step in our work includes the evaluation of the influence of smoke plume height on the atmospheric composition over the southern hemisphere, based on insights from the analysis of the smoke plume climatology across the Amazon, and further application of this approach to other geographic regions.

Chapter 4

Biomass burning influence on CO and ozone over the Amazon: sensitivity to vertical smoke distribution and source contributions [†]

4.1 Introduction

Across the Amazon basin, with an area of $6,300,000 \text{ km}^2$ and a population of 25 million people (Davidson et al., 2012) distributed in eight countries, millions of hectares burn every year. Biomass burning in the Amazon is an important global source of emissions, which contributes by 15% to the global total fire emissions (Van der Werf et al., 2010) and drives the interannual variability in the southern

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hemisphere (SH) atmospheric composition (Thompson et al., 2001, Edwards et al., 2003, 2006). Emissions from biomass burning include large amounts of aerosols, greenhouse gases and reactive trace gases, i.e., precursors of ozone to the atmosphere, such as CO and NO_x (Andreae and Merlet, 2001). Moreover, biomass burning is believed to be the most important source of CO and NO_x in the tropics (Chameides et al., 1992, Crutzen and Carmichael, 1993), which can lead to enhancements in global tropospheric ozone of around 3–5% (Ziemke et al., 2009). Specifically across the Amazon, biomass burning is responsible for increases in surface ozone of around 30–40% during the fire season (Galanter et al., 2000).

Tropospheric O_3 is a secondary product from biomass burning, an important greenhouse gas (IPCC, 2007) and air pollutant, harmful to human health (e.g., Levy et al., 2001, Ito et al., 2005, Bell et al., 2006) and detrimental to crops and plants (e.g., Sitch et al., 2007, Van Dingenen et al., 2009a). Inhalation of elevated levels of ozone can decrease lung function, aggravate asthma, cause higher susceptibility to respiratory infections and premature death (e.g., Bell et al., 2004, 2006, Kheirbek et al., 2013, Liu et al., 2018). High and long exposure to ozone concentrations can also damage leaf tissue, decrease net productivity of plants and reduce crop yields (e.g., Reich and Amundson, 1985, Avnery et al., 2011, Ghude et al., 2014, Yue and Unger, 2018). Ozone forms as a result of photochemical reactions of many combustion products in the atmosphere, i.e., carbon monoxide (CO), volatile organic compounds (VOC) and nitrogen oxides $(NO_x = NO + NO_2)$. These reactions take place within a few tens of minutes after released (Goode et al., 2000, Yokelson et al., 2003, Jaffe and Wigder, 2012). The Amazon is strongly impacted by emissions of these products, from biomass burning and vegetation (Galanter et al., 2000, Edwards) et al., 2006, Gloudemans et al., 2006, Williams et al., 2013), particularly during the burning season.

During the Amazon burning season, previous studies have reported high ozone mixing ratios (60–80 ppb) in the middle and upper troposphere over the region (e.g., Andreae et al., 1988, Kirchhoff et al., 1992, Fishman et al., 1996b, Betts et al.,
2002). Important enhancements on surface ozone (>20-40 ppb), with daily maxima of 100 ppb have also been reported (e.g., Kirchhoff et al., 1992, Kirkman et al., 2002, Artaxo et al., 2005, Bela et al., 2015). Furthermore, high O_3 enhancements have been generally observed particularly in tropical/subtropical aged plumes, as a result of a more efficient O_3 production due to the greater flux of solar radiation and higher temperatures (Andreae et al., 1994, Jaffe and Wigder, 2012). With a lifetime of several weeks, O₃ can be transported long-range downwind or be produced downwind after long-range transport of its precursors (Jaffe et al., 2004, Real et al., 2007, Nassar et al., 2009), which can produce exceedances of the O_3 air quality standards in metropolitan areas far from the fires (Jaffe et al., 2008, Pfister et al., 2008, Chalbot et al., 2013, Rubio et al., 2015, Brey and Fischer, 2016). Despite the high O₃ levels found every year across the Amazon region, little is known about the impact on air quality. For instance, Pacifico et al. (2015) modelled the impact of fire-induced O_3 damage on the Amazonian forest, and estimated mean reductions in net productivity of 15% and up to 60% at certain grids. However, models over the Amazon, including the one used in Pacifico et al. (2015), tend to overestimate O_3 levels within the boundary layer, specifically under clean air conditions, and underestimate elevated O_3 (Pacifico et al., 2015, Bela et al., 2015), as a result of poor representation of biomass burning and the altitude to which biomass burning emissions are injected in the atmosphere i.e., the injection height (Andreae et al., 2012), among other factors.

The injection height is one of the main sources of uncertainty in chemical transport models (CTM). It determines the lifetime and downwind transport of pollutants, and thus, the magnitude of their impact (Paugam et al., 2016). Most large-scale CTMs used a simple approach to represent smoke injection heights: some models release all biomass burning emissions at the surface, others distributed evenly within the boundary layer (Bey et al., 2001) or homogeneously distributed from the surface to a prescribed height (Dentener et al., 2006). Results from models that inject a certain fraction of smoke in the free troposphere (FT) have shown improvements in their estimates of surface and total column CO downwind of the fires (Leung et al., 2007, Turquety et al., 2007). Some modelling studies have used complex dynamical parametrisations based on thermal convective approaches for pyroconvection embedded on CTMs (Freitas et al., 2007, Rio et al., 2010). They showed that injecting fire emissions in the FT has a large impact on CO concentrations downwind (Freitas et al., 2006, 2007), but showed little agreement with observations for the elevated numerical cost (Val Martin et al., 2012) and complexity that can result in errors (Ichoku et al., 2012). Over the Amazon, results obtained using complex dynamical parametrisations tend to overestimate smoke plume heights (Freitas et al., 2007, Archer-Nicholls et al., 2015).

The injection height is highly variable and depends on the climatic zone, the season, the type of biome burned, the size and intensity of the fire and the atmospheric conditions (e.g., Amiridis et al., 2010, Val Martin et al., 2010, 2018b). Specifically over the Amazon, Gonzalez-Alonso et al. (2019), based on a comprehensive 8-year climatology of smoke plume heights derived from satellite observations from MISR and CALIOP, reported large variability on smoke plume heights, from a few meters to 6 km, with most smoke concentrated below 2.5 km. Their results agree well with previous studies across the region (Baars et al., 2012, Marenco et al., 2016). However, those studies were limited to a specific location or flight tracks. Gonzalez-Alonso et al. (2019) also showed the importance of drought conditions on the injection heights and aerosol loading over the region. Drought tends to favour smouldering fires, which are associated with low smoke injection heights and larger smoke emissions, and this, in turn, has important implications for air quality. Recently, results from a modelling study that applied a novel global injection height scheme derived from MISR observations (Val Martin et al., 2018b), which included smoke plume heights from Gonzalez-Alonso et al. (2019), showed an improvement in near-source surface concentrations and vertical profiles of CO over the Amazon (Zhu et al., 2018).

Future warming climate is expected to enhance frequency of droughts and fire

activity (Aragao et al., 2008, Li et al., 2008, Spracklen et al., 2009, Cochrane and Barber, 2009), as well as intensify biogenic emissions (Heald et al., 2008), which may result in an increase in O_3 precursor emissions. This, in combination with rapid urbanization, could pose a potential risk for millions of people's health and vegetation across the Amazon. Given the large contribution of Amazonian biomass burning emissions to O_3 levels, it is important to understand the distribution of fire pollution across the region, as well as its impacts on surface O_3 levels. Here, we aim at improving the representation on the vertical distribution of biomass burning emissions in the Amazon and assessing the impacts on surface ozone levels across the region, with implications for air quality. To our knowledge, this is the first time that a study uses an improved representation of smoke injection height across the Amazon to address the relative impact of fire pollution on O_3 air quality. We use a global earth system model (ESM) with the Val Martin et al. (2018b) injection height parametrisation, which includes observations of Amazonian smoke plume heights, and evaluate the simulation with ozonesondes, satellite observations, and ground-based and aircraft measurements. Then, we assess the impact of biomass burning emissions from the Amazon on surface ozone, with focus on the associated exceedances of the ozone critical levels for air quality. Section 2 of this paper describes the global ESM and simulation experiments and Section 3 introduces the observational dataset used. Section 4 presents the model evaluation and Section 5 quantifies the impact of fire-induced O_3 on air quality.

4.2 Modelling Framework

To assess the performance of the updated representation of the vertical distribution of biomass burning emissions over the Amazon, and study the implication to simulated surface ozone, we use the Community Atmosphere Model with Chemistry (CAM6-Chem), a global atmospheric chemical transport model, which is part of the ESM Community Earth System Model (CESM). This section presents briefly the model and describes the planned modelling simulations.

4.2.1 Model description

We use version 2 of the Community Earth System Model (CESM; Lamarque et al. (2012)) developed at the National Center for Atmospheric Research (NCAR). CESM is a fully coupled Earth system model (ESM), which consists of seven geophysical model components, the Community Atmosphere Model Version 6 (CAM6; Bogenschutz et al. (2018)), the Community Land Model Version 5.0 (CLM5; Lawrence et al. (2018)), and the river, ocean-wave, ocean, land-ice and sea-ice components. In this study, we run CESM2 with only the atmosphere component with chemistry (CAM6-chem) coupled with CLM5, and with prescribed sea-surface temperatures (SST) and sea-ice fractions (Hurrell et al., 2008). This setting allows for instantaneous flux exchanges between the land and the atmosphere, including biogenic emissions and dry deposition of gases and aerosols (Oleson et al., 2010). We refer to the model as CAM6-chem hereafter. All simulations are performed at a horizontal resolution of 0.9°latitude x 1.25°longitude and vertical resolution of 56 hybrid vertical sigma levels (top around 40 km), with a time step of 30 minutes.

CAM6-Chem includes over 200 species and 400 reactions (Tilmes et al., 2016), based on MOZART-4 (Emmons et al., 2010). Aerosols treatment follows the fourmode version of the modal aerosol module (MAM4) parametrisation, which differs four modes of aerosols size and distribution: coarse, aitken, accumulation mode and primary carbon (Liu et al., 2016). We use prescribed meteorology from the Modern-Era Retrospective Analysis for Research and Applications (MERRA-2) (Bosilovich et al., 2015). Table 4.1 shows a summary of several key species emissions used in this work. Anthropogenic emissions are monthly averages and extracted from the Coupled Model Intercomparison Project Phase 6 (CMIP6) (Eyring et al., 2016), which uses the Community Emissions Data System (CEDS) (Hoesly et al., 2018). Biogenic emissions are interactively computed by the Model of Emissions and Aerosols from Nature (MEGAN), version 2.1 (Guenther et al., 2012), integrated into the CLM5. MEGAN 2.1 estimates volatile organic compounds based on emission factors, plant functional type, leaf area index (LAI), leaf age, soil moisture, light and temperature. We include emissions from volcanoes and aircraft vertically distributed, from Dentener et al. (2006) and Lamarque et al. (2010), respectively. Emissions of NO_x from lightning follow Emmons et al. (2010) approach, that uses the Price parametrisation (Price and Rind, 1992, Price et al., 1997). These emissions are scaled to a global annual rate of 3-4 Tg(N)/year and vertically distributed following DeCaria et al. (2005).

Global $[Tg]$	CO	NO_x	BC	OC	Isoprene
Anthropogenic	576.09	81.26	7.85	27.20	-
Biogenic	75.54	-	-	-	479.24
Biomass burning	373.04	18.46	2.05	31.19	0.79
Amazon [Tg]	CO	NO_x	BC	OC	Isoprene
Anthropogenic	38.32	4.47	0.42	1.72	-
Diogenia	00.04				000.05
Diogenic	28.04	-	-	-	230.35

Table 4.1: Summary of the global and domain-based emissions from main sectors for 2012.

Biomass burning emissions are obtained from the Fire Inventory from NCAR version 1.5 (FINNv1.5; Wiedinmyer et al. (2011)), which provides global trace gases and particles emissions from biomass burning, at high temporal (daily) and spatial resolution (1 km) from January 2002. FINNv1.5 emissions are calculated via satellite observations of active fires and land cover, estimates of fuel loadings and emission factors for open burning compiled by Akagi et al. (2011) and Andreae and Merlet (2001).

4.2.2 Biomass burning injection height parametrisation

We include a novel global biomass burning injection height scheme (Val Martin et al., 2018b) to distribute FINN's emissions in altitude. The scheme assigns fire emission fractions at different altitudes depending on land cover, fire region and season, from

0 to 8 km and at a vertical resolution of 250 m. The plume height parametrisation is based on a statistical analysis of global MISR smoke plume heights observations for 2008–2010, compiled as part of the MISR Plume Height Project2 (MPHP2) for the AeroCom multi-model biomass burning experiment. For a detailed description of the plume height parametrisation, see Val Martin et al. (2018b).

To vertically distribute FINN fire emissions into CAM6-Chem, we first identify the region and assign plant functional types per grid. For each grid, depending on the region that contains it, the fraction of plant functional types and month of the year, a specific emission profile is calculated and applied. As an example, Figure 4.1 shows the summertime vertical distribution of fire emissions across the main biomes in the Amazon. An overview of the MISR instrument, standard products and the MISR plume digitizing tool is given in Chapter 2. The MISR plume database can be found in Val Martin et al. (2018b).



Figure 4.1: Vertical distribution of biomass burning emissions, in August, for the main biomes across the Amazon region.

4.2.3 Experimental setup

To assess the performance of the smoke injection height parametrisation over the Amazon, we conduct two simulations: a control simulation with biomass burning emissions released at the surface level (CAMsurf), and a simulation with biomass burning emissions vertically distributed following Val Martin et al. (2018b) scheme (CAMvert). Both simulations run from 2007-2012 with one year spin up. The simulated time frame coincides with recent aircraft campaigns over the region, which provide CO profiles to validate our results at high spatial and temporal resolution. In addition, we aim at investigating the contribution of biomass burning in the Amazon to surface ozone levels. For that, we perform an additional simulation for 2011-212 with the same configuration as CAMvert (i.e., biomass burning emissions vertically distributed) but with Amazon fire emissions set to zero (CAMzeroBB).

4.2.4 CO Tags

We introduce global tags of emitted CO associated with specific sources following the work of Emmons et al. (2010) and Gaubert et al. (2016). The CO tags allow us to assess the influence from a particular source, region or biome on the total CO levels and quantify the relative importance of each source. These CO tags are subject to the same reactions as the total CO (Emmons et al., 2010, Gaubert et al., 2016) and include CO emitted by the main primary sources, i.e., anthropogenic, biomass burning, oceanic and biogenic. CO produced from chemical oxidation represents the secondary source of CO, and the sum of primary and secondary CO is the total CO. Additionally, we include CO tags for the main biomass burning regions across the globe, i.e., Europe, Boreal North America, Boreal Asia, Temperate North America, Central East-Asia, South East-Asia, Equatorial Asia, India, Australia, South America, Central America, North Africa, South Africa and Amazon (Figure B1 in Appendix B), and for the main biomes in the Amazon, i.e., tropical forest, savanna, grassland and cropland. For the latter, we associate FINN emissions to biome type using MODIS MCD12C1 Land Cover product (Friedl et al., 2010) and select only the biome specific to the Amazon: tropical forest, savanna and others (agricultural and grassland).

4.3 Observational datasets for model evaluation

Simulations performed with CAM6-chem have been extensively evaluated against satellite, ozonesondes, aircraft and ground observations of key pollutants on a global and regional scale (e.g., Lamarque et al., 2012, Tilmes et al., 2015, Val Martin et al., 2015, Tilmes et al., 2016). Here we focus our evaluation on carbon monoxide (CO) and ozone (O₃) over the Amazon domain [90°–10°W Longitude, 20°S–10°N Latitude], using aircraft campaigns and CO retrievals from the Measurement of Pollution in the Troposphere (MOPITT), and ozonesondes and long-term surface measurements of O₃.

4.3.1 CO Observations

CO is one of the main gases emitted by biomass burning and an important ozone precursor, whose relatively long lifetime (weeks to months) make it an excellent tracer for fire emissions (Edwards et al., 2006, Yurganov et al., 2010, Gatti et al., 2014).

We use CO vertical profiles measured from three aircraft campaigns to assess CAMvert versus CAMsurf performance. These aircraft campaigns include the Regional Carbon Balance in Amazonia (BARCA Balanco Atmosferico Regional de Carbono na Amazonia), the Gatti et al. (2014) flights and the South AMerican Biomass Burning Analyses (SAMBBA). The dataset allows us to evaluate the impact of the smoke plume injection heights in different years and conditions. BARCA emerged as a combination of observations and analysis framework to quantify basin-scale carbon fluxes with greenhouse gases and aerosols measurements over the Amazon (Andreae et al., 2012). BARCA was divided into two phases that represent the shift in the atmospheric conditions during the two seasons: BARCA-A, at the end of the dry season in 2008 (16 November–2 December), and BARCA–B, at the end of the wet season in 2009 (15–28 May). Flights in BARCA covered an altitude range from the surface to about 4500 m over most of the Amazon Basin. Gatti et al. (2014) used CO measurements as part of a sampling programme to observe climate sensitivity of the Amazon carbon pools during 2010 and 2011. The project consisted of bi-weekly vertical profiles of CO at four sites across the Amazon: Alta Floresta (ALF; 8.80°S, 56.75°W), Rio Branco (RBA; 9.38°S, 67.62°W), Santarem (SAN; 2.86°S, 54.95°W) and Tabatinga (TAB; 5.96°S, 70.06°W). Aircraft measurements were taken descending in spiral from approximately 4420 m to 30 m above sea level from 12:00 to 13:00 local time (LT). SAMBBA consisted of a combination of remote sensing, groundbased and aircraft measurements, which aimed to investigate the impacts of biomass burning pollution over South America (Allan et al., 2014). SAMBBA was conducted during the dry season of 2012 (14 September–3 October) and included 20 scientific flights sampling the Amazonian atmosphere from the surface up to almost 8 km.

MOPITT is a space-borne instrument aboard the NASA EOS Terra satellite designed to study the distribution, transport, sources, and sinks of CO in the troposphere. It uses gas correlation spectroscopy to retrieve measurements of the emitted and reflected radiance from the Earth in three spectral bands. The amount of energy absorbed into the sensor is correlated to the presence of CO in the atmosphere. MOPITT has a swath of 640 km and a spatial resolution of 22 km at nadir, and provides long record of global CO (since 2000) and global coverage every 3 days (Deeter et al., 2003).

We use daytime monthly joint retrievals (Thermal infra-red TIR and near infrared NIR product) of the Level 3 Version 7, which provide total CO column and CO profiles in ten levels, from the surface to 100 hPa (Deeter et al., 2003, 2014, 2018). The use of combined TIR and NIR retrievals improves the sensitivity to CO in the lower troposphere, particularly at daytime over land. To compare the MOPITT retrievals to our monthly CO simulations, we interpolate the model outputs to the ten-level pressure (surface-100 hPa) and horizontal 1°x 1°MOPITT grid. Then, we apply the a priori and averaging kernels included in the MOPITT retrievals to account for the sensitivity of the retrieval to the truly observed profiles (Morgenstern et al., 2012).

4.3.2 Ozone Observations

We use ozonesonde observations compiled by Tilmes et al. (2012) to assess the performance of the simulated ozone vertical distributions. The dataset includes vertical ozone monthly profiles for 42 stations from 3 networks (World Ozone and Ultraviolet Data Center (WOUDC), the NOAA Earth System Research Laboratory (ESRL) and the Southern Hemisphere ADditional OZonesondes (SHADOZ)) for the period 1995–2011, and also regionally aggregated by similar ozone characteristics for their seasonal median and shape of pdfs. We focus on two regions: Equatorial Americas and Atlantic/Africa, which contain two and three stations, respectively (Figure B2 in Appendix B; Tilmes et al. (2012)). For the comparison, simulated ozone monthly outputs were first interpolated to the specific sondes locations and then aggregated and averaged by region and period.

In addition, we assess the ability of our simulation experiments to capture the spatial and temporal distribution of surface ozone. We evaluate our results with ground-based ozone measured at eight stations located within or close to the Amazon, from the Tropospheric Ozone Assessment Report (TOAR; Schultz et al. (2017)). These data are publicly available and free to download through PANGAEA. We use a minimum of two-years monthly means of the records available and coincident with the simulations period (2007–2012). (Figure B2 and Table B1). Simulated monthly ozone mixing ratios at the surface are interpolated at the location of the stations and averaged for the run period.

4.4 Model performance with the smoke injection height parametrisation

We first compare results from CAMvert and CAMsurf to assess the impact of the biomass burning injection height distribution on the vertical distribution of CO and O_3 across the Amazon. We focus on differences between CAMvert and CAMsurf during March and September 2012, as these months represent wet versus dry conditions, and low (March) versus high (September) fire activity (Figure B3). Secondly, we evaluate simulated CO and O_3 from CAMsurf and CAMvert against observations to assess the performance of the simulations on representing CO and O_3 across the Amazon domain.

4.4.1 Impact of the smoke injection height scheme on simulated CO

Figure 4.2 shows the relative changes in the simulated CO mixing ratios between CAMvert and CAMsurf at the surface, 2 km, 6 km and the column from the surface to 8 km. Figure B4 shows the CO mixing ratios and column (0–8 km) value for these simulations. We choose 8 km as the top height in our analysis because smoke over the Amazon is rarely found above this altitude (Gonzalez-Alonso et al., 2019). Furthermore, the maximum altitude at which fire emissions are distributed over this region is typically 6 km (Val Martin et al., 2018b). As expected, the new injection scheme (CAMvert) decreases simulated CO mixing ratios at the surface, and increases them in the free troposphere, at 2 km and 6 km altitudes, especially near the fires in September. For example, simulated CO mixing ratios in CAMvert are reduced by around 70% (\sim 800 ppb) at the surface, increased by 30% (\sim 50 ppb) at 2 km, and by 7% (3 ppb) at 6 km above the fires, compared to CAMsurf in September. Small or negligible changes in simulated CO are found during the low fire active month of March. We find that the new injection height scheme

has important effects on the simulated CO column (0-8 km) on a regional scale (Figure 4.2b), especially at the peak of the biomass burning season in September. Simulated CO column is increased (5-10%) across the Peruvian coast, the Atlantic Ocean and the eastern Amazon, and decreased (10-15%) right above the fires at the arc of the deforestation. Previous studies have also shown that injecting fire emissions decreases CO in the source regions and increases it downwind, as CO lifetime is typically longer in the FT (Chen et al., 2009). Our results are consistent with the effect that injecting fire emissions at higher altitudes has on pollutant lifetime and downwind transport.

Evaluation of simulated CO with aircraft observations

We evaluate simulated CO from CAMvert and CAMsurf near the fire source with vertical profiles of CO from measurements gathered in 159 flights during three recent aircraft campaigns in the Amazon. We limit our comparison to the flights during the burning season (July-September) to maximize the differences in the simulated CO profiles in CAMvert and CAMsurf from the fire injection scheme. Figure 4.3 shows an example of simulated and observed CO from a flight during BARCA-A and SAMBBA, and the averaged CO observations in RBA site from Gatti et al. (2014) during 2010. We choose that SAMBBA flight and RBA site because they are highly influenced by biomass burning emissions (Brito et al., 2014, Gatti et al., 2014). For BARCA-A, we select a vertical profile where both simulations represent fairly well the CO vertical distribution, and focus on the differences from the injection height scheme. During BARCA-A aircraft measurements were only influenced by biomass burning during two days (Andreae et al., 2012) and our two simulations fail at capturing the observed high CO mixing ratios (400 ppb; Andreae et al. (2012)) at those two flights, due to underestimated CO emissions in FINN. Consistent with Section 4.4, our comparison for SAMBBA and RBA shows that CO mixing ratios in CAMvert are significantly reduced below 2 km (by up to 150 ppb) and enhanced from around 2–4 km (up to 40 ppb). In general, CAMvert matches better the



Figure 4.2: Relative differences (%) [(CAMvert – CAMsurf)/CAMsurf] in simulated CO mixing ratios between CAMvert and CAMsurf at three different levels: the surface, 2 km and 6 km (a), and the column (0-8 km) (b), for March and September 2012. Absolute mixing ratios (ppb) and column of CO (molec/cm²) are shown in Figure B4, in Appendix B.

observations at most levels during smoke-impacted conditions than CAMsurf. For the BARCA-A flight, CAMvert matches better the observations, specifically at 1– 2 km, although this flight was hardly impacted by biomass burning compared to the SAMBBA and RBA site (averaged CO mixing ratio is 160 ppb, compared to 360 ppb at SAMBBA and 220 ppb at RBA site).

We also compare our simulated CO mixing ratio profiles from CAMvert and CAMsurf with other SAMBBA and BARCA flights and other Gatti et al. (2014) sites (TAB, ALF, and SAN), and the impact of new injection scheme on CO was low for some of them. Consistent with Zhu et al. (2018), we find that the evaluation of simulated CO against aircraft measurements is not that straightforward. Across the Amazon, fire emissions are highly variable and depend on many factors i.e., topography, type of vegetation burned, season, weather conditions and regional droughts on a particular year (Van der Werf et al., 2010). In general, FINN tends to overestimate fire emissions during the burning season across some regions in the Amazon (Pereira et al., 2016). In our analysis, a considerable number of simulated CO profiles had a poor agreement with the observations, as the model was not able to capture the extreme high CO resulting from high biomass burning influence or tended to overestimate CO under low biomass burning influence. Another aspect to consider is that the MISR injection height scheme may be biased low, as it is based on a statistical analysis of smoke plume heights observed during the early morning (10:00–11:00 LT) (Val Martin et al., 2018b). Most of the flights were performed in the afternoon (Andreae et al., 2012, Gatti et al., 2014), when the PBL is fully developed and fires are more energetic, and thus, smoke heights are higher, as shown for the Amazon fires by Gonzalez-Alonso et al. (2019).

Evaluation of simulated CO with MOPITT

We use monthly CO retrievals from MOPITT to evaluate simulated CO from CAMvert and CAMsurf at a larger scale. Figure 4.4 presents CO total column from MOPITT, CAMvert and CAMsurf in September 2012 over our Amazon domain. CAMvert and CAMsurf simulations are convolved with the MOPITT a priori and averaging kernels for comparison. Absolute differences between simulated and MO-PITT CO total column are also included in Figure 4.4. MOPITT shows high levels of CO across the arc of deforestation and efficient transport of CO from Africa



Figure 4.3: Examples of vertical profiles of CO mixing ratios from observations (black), simulated with CAM-vert (blue) and CAMsurf (red) for BARCA-A (a), SAMMBA (b) and RBA (c). Shown are median for SAMMBA and BARCA-A and average for RBA, with the horizontal bars indicating the 10th and 90th percentiles.

across the Atlantic, as reported by previous MOPITT studies (Edwards et al., 2006). Our comparison shows important biases between both CAM6-chem simulations and MOPITT. CAM6-chem misses the transport out of Africa, overestimates total CO column over western Amazon and along the Andes, and underestimates it over Eastern Amazon (by $\sim 30\%$). We notice a small difference between CAMvert and CAMsurf simulated CO total column with respect to MOPITT in this relatively large scale analysis, and compare them at two vertical levels in Figure 4.5. We display the relative bias of simulated CO mixing ratios against MOPITT CO retrieved at 400 and 800 hPa (about 7 and 2 km altitude, respectively) for September 2012. Figure B5 displays the comparison for March 2012. These two levels represent the lower troposphere (LT) and upper troposphere (UT) that include most of the information from the retrieval with the least contribution from the a priori (Deeter et al., 2018). We find that both CAMvert and CAMsurf overestimate mean CO mixing ratios, in particular at 800 hPa over western Amazon, with a relatively large bias up to 80–120% in March and September. Contrariwise, both simulations underestimate CO across the Eastern Amazon, in particular at 400 hPa with relative biases up to 60-80% in March and September.

Deeter et al. (2018) showed that MOPITT CO retrievals have a negative mean bias across the Amazon of about -16% and up to -27% due to the low sensitivity of MOPITT to the LT. This negative bias does not fully explain the positive bias we find in our comparison. Previous studies have also detected similar discrepancies between CAM6-chem and MOPITT (e.g., Zeng et al., 2015). They suggested that biases may be caused by poor representation of fire emissions, as well as model atmospheric processes (i.e., convection), which may result in accumulation of CO from fires along the Andes that act as a barrier during its transport by the easterly winds. The large bias between CAM6-chem and MOPITT makes it difficult to assess the large scale effect of including an injection height scheme in our model. Furthermore, most of the biomass burning emissions in CAMvert are released in the first 3 km, where MOPITT has the lowest sensitivity to CO, therefore, comparison



Figure 4.4: Mean MOPITT, CAMsurf and CAMvert CO total columns for September 2012 (top) and CAMsurf and CAMvert bias with respect to MOPITT (bottom). Red squares divide the domain in three fire regions North (90°–10°W Longitude, 5°–20°N Latitude), South-West (90°–50°W, 30°S–5°N) and South-East (50°–10°W, 30°S–5°N)(bottom left).

of CAMvert and CAMsurf differences against MOPITT does not provide much information. However, our analysis shows that during the burning season, the bias of CAMvert with respect to MOPITT is slightly higher than in CAMsurf at 800 hPa, where CAMvert injects most of the CO (12% versus 10%, respectively). Moreover, CAMvert improves somewhat the low bias observed at 400 hPa, being 17% versus 18% in CAMsurf.

To elucidate a regional and seasonal effect on our CAM-chem-MOPITT evaluation, Figure 4.6 compares monthly CO total column across three fire-influenced regions: north, southeast and southwest (depicted in Figure 4.4). We also include results from our simulation with Amazon biomass burning set to zero (CAMzerobb) to have a reference of the impact of biomass burning across these regions. This analysis confirms again the persistent low bias on the simulated CO across most of the Amazon, in particular north and southeast (15–20%), and positive bias during the biomass burning season in southwest (~5%). However, CAMvert and CAMsurf show an acceptable annual low bias of ~15% compared to MOPITT total CO column (not



Figure 4.5: Averaged MOPITT CO mixing ratios and relative bias (in %) of CAMsurf and CAMvert with respect to MOPITT at 400 and 800 hPa for September 2012.

shown). Also, CAMvert slightly improves CO total column compared to MOPITT at the peak of the burning season (August-October) in the north and southeastern regions by 1-2%. Previous studies have also found small differences when comparing model simulations with and without injection heights against large-scale observations with low vertical information (e.g., MOPITT) (e.g., Chen et al., 2009, Zhu et al., 2018). As in Zhu et al. (2018), we find better agreement in simulated CO with the improved vertical injection height scheme (CAMvert) when the model is compared to specific plumes from aircraft measurements and these improved simulations can have important implications for air quality.

4.4.2 Impact of smoke injection height scheme on simulated O_3

Figure 4.7 displays the relative changes in the simulated O_3 mixing ratios between CAMvert and CAMsurf at the surface, 2 km, 6 km and the column from the surface to 8 km. Figure B6 shows the simulated O_3 mixing ratios and column (0–8 km) values. Injecting O_3 precursors at higher altitudes leads to changes in the formation and vertical distribution of O_3 particularly near the source during the burning sea-



Figure 4.6: Monthly averages of CO total columns for MOPITT (black), CAMsurf (red), CAMvert (blue) and CAMzeroBB (green) for three major regions in our domain (see Figure 4.4) in 2012.

son. It decreases simulated O_3 mixing ratios at the surface, and increases them in the low-mid troposphere. For instance, in September, simulated O_3 mixing ratios in CAMvert are reduced by 40% (~25 ppb) at the surface and increased by 10% (~5 ppb) at 2 km above the fires. At 6 km, the sensitivity of O_3 to the injection height scheme above the fires is minimal. Also, we observe little change above the fires in March. On a regional scale, the new injection height scheme (CAMvert) decreases the simulated O_3 column (0–8 km) across the source regions i.e., the arc of deforestation (Figure 4.7b) (~5%) in September. O_3 formation is complex and many factors are involved i.e., availability of NO_x , the limiting factor in O_3 production in the Amazon. Moreover, we observe NO_x column reductions across the source regions (by 25%) (not shown) coincident with the O_3 reductions. In addition, reductions in CAMvert O_3 column may be partly caused because injecting biomass burning aerosols at higher altitudes, may decrease ozone formation via photolysis attenuation produced from scattering or absorbing solar radiation by aerosols (Reid et al., 2005, Real et al., 2007).

Evaluation of simulated O_3 with ozonesondes

We use monthly mean profiles from ozonesondes averaged in two regions to evaluate simulated O₃ from CAMvert and CAMsurf at a larger scale. Figure 4.8 shows the simulated and observed O₃ mixing ratios at 800, 600 and 400 hPa (~2, 4 and 7 km) for the Equatorial and African/Atlantic regions. Overall, CAM6-chem captures increasing O₃ mixing ratios with height and represent quite well O₃ spatial and temporal distribution, particularly at 800 and 600 hPa. The simulated O₃ mixing ratios are in the uncertainty limits of the measurements, although we find some bias. CAM6-chem overestimates O₃ in the Equatorial region at 800 hPa (25%) and at 600 hPa (< 10%) during the Amazon burning season, probably caused by poor representation of O₃ precursor emissions by FINN. Over the Atlantic, from July to December, CAM6-chem misses large O₃ mixing ratios (~ 35%) commonly found every year (Thompson et al., 2001). At 400 hPa, the CAM6-chem simulations show





n simulated O_3 mixing ratios]Relative differences (%) in simulated O_3 mixing ratios between CAMvert and CAMsurf at three different levels: the surface, 2 km and 6 km (a), and the column (0-8 km) (b), for March and September 2012. Absolute mixing ratios (ppb) and column of O_3 (DU) are shown in Figure B6.

a poorer representation of O_3 mixing ratios than at the rest of levels. Previous modelling studies reported a systematic low bias in the SH, particularly in the UT, and argued that it is associated with missing recirculated pollution from southeastern Brazil, biomass burning and/or lightning NO_x production (e.g., Bela et al., 2015,



Young et al., 2018, Ziemke et al., 2009).

Figure 4.8: Monthly mean O_3 mixing ratios averaged across the Equatorial and Atlantic/African regions at 800, 600 and 400 hPa for ozonesonde observations (black), CAMsurf (red) and CAMvert (blue). Error bars show the standard deviation of the mean.

CAMvert slightly improves O_3 mixing ratios in the Equatorial region, particularly at 600 hPa and during the burning season. Over the Atlantic, CAMsurf gives slightly higher O_3 mixing ratios, and represents somehow better O_3 mixing ratios, where CAM6-chem shows a low bias. As in the evaluation with MOPITT, in Section 4.4.1, we notice small differences in the simulated O_3 mixing ratios between CAMsurf and CAMvert when compared against large-scale measurements with low vertical information. However, at smaller scales, injection heights can considerably impact surface O_3 levels. We investigate this below.

Evaluation of simulated O_3 with surface ozone measurements

We use long-term measurements from ground-based TOAR stations to evaluate CAMvert and CAMsurf performance at representing the spatial and temporal distribution of surface ozone. Figures 4.9 and B7 show the observed and simulated monthly mean surface O_3 mixing ratios from the TOAR stations, CAMvert, CAMsurf and CAMzerobb. We include CAMzeroBB as a reference to the total impact of biomass burning emissions on surface ozone. Observations of monthly mean surface O_3 range from few ppb to 40 ppb, depending on location and month. During the wet season, TOAR measurements in the Amazon basin (Amazon TT34 and Porto Velho), are lower than 10 ppb, evidencing the low influence of polluted air across the Amazon. However, during the burning season, O_3 mixing ratios at these stations increase up to almost 20 ppb. Surface O_3 levels in central Brazil can be 2–3 times higher than background concentrations during periods of intense burning (Kirchhoff et al., 1989). In general, the CAM6-chem captures well the seasonality of the simulated O_3 monthly means, although with some persistent high bias (6–16 ppb). Similar biases were previously documented across the Amazon (5–15 ppb), especially under clean air conditions, and were suggested to arise for many reasons i.e., model resolution, uncertainties in fire emissions, O_3 dry deposition scheme in the models, excessive sensitivity to NO_x emissions, lightning NO_x production and transport of O_3 and its precursors across the area (Pacifico et al., 2015, Bela et al., 2015).

Differences in the simulated surface O_3 mixing ratios among the three simulations are large at the sites impacted by biomass burning, as it is the case of Amazon TT34 and Porto Velho, from August to November. Therefore, we focus the evaluation at these stations (Figure 4.9). CAMvert reduces surface O_3 mixing ratios compared to CAMsurf. CAMvert reductions range from 1.16% during the wet season, to over 100% in August, with a mean reduction in surface O_3 of around 70% during the burning season (July-November). This is consistent with Section 4.4.2. The Amazon TT34 and Porto Velho stations are located in the arc of deforestation, where we observe the largest impact of the injection heights on surface O_3 in September, with up to 55% of reduction in CAMvert at certain grids (Figure 4.7). CAMvert improves surface O_3 mixing ratios with respect to TOAR measurements across the Amazon during the burning season, particularly at the Amazon TT34 station. At



Figure 4.9: Monthly averages of O_3 mixing ratios at the Amazon TT34 and Porto Velho surface sites from TOAR observations (black), CAMvert (blue), CAMsurf (red) and CAMzeroBB (green). Vertical bars indicate the standard deviation of the mean.

Porto Velho, both simulations overestimate surface O_3 , which may be related to the uncertainty in the measurements because of the low availability of data at the site. However, CAMvert still matches significantly better the observations. The evaluation presented here demonstrates the large impact of the improved injection height scheme on surface O_3 when compared at a local scale, important for air quality assessment.

4.5 Large scale impacts of biomass burning on CO and O_3

In this section, we present an analysis of the impact of biomass burning on CO and O_3 with simulations from CAMvert.

4.5.1 Source attribution of CO

We use tags of emitted CO from different sources to quantify the contribution of biomass burning emissions to the total CO in the region and the relative importance of biomass burning compared to other sources. Figure 4.10 presents the monthly relative contributions of the CO tags in 2012 for major emission sources (i.e., anthropogenic, biomass burning, oceans and biogenic), for main biomass burning regions that may directly impact the Amazon (i.e., Amazon, NH Africa, SH Africa, Central America and South America), and for main biomass burning sources within the Amazon (i.e., tropical forest, savanna and other). Firstly, we estimate that primary sources contribute to about 37% of the total CO over the Amazon, and secondary sources, i.e. due to oxidation of biogenic products, contribute to about 67%. This estimate is consistent with previous studies that showed secondary production as the dominant source of CO in the SH (Zeng et al., 2015), mainly due to oxidation of isoprene (Stein et al., 2014), which is a major emission within our domain (Table 4.1).

For primary CO (Figure 4.10a), our analysis shows that anthropogenic emissions are the major source of CO from January to August (44–52%), whereas biomass burning dominates from September to December (48-65%), with its peak in October. On an annual basis, both sources contribute equally to the total CO, with an average of about 42%. Biogenic sources contribute about 8–15% and ocean emissions about 2-3%.

The main source of CO from biomass burning is local fire emissions from Septem-



Figure 4.10: Monthly relative contributions to primary CO for major emission sources a), for main biomass burning regions b) and for main biomes in the Amazon c), for 2012.

ber to January (48–79%), whereas contributions from NH African fires are important from February to June (55–78%) and from SH African fires from July to September (46–64%). Central and South American fires have little influence (<15%) on the total CO from fires across our region. Overall, we find that contribution from African fires (24 and 35%) can be as significant as Amazonian fires (36%) in our domain. Biomass burning emissions in Africa are responsible for half of the global carbon emissions (Van der Werf et al., 2010) and long-range transport from biomass burning in Africa has been shown to have a strong influence over South America (Edwards et al., 2006). We also find some interannual variability in the relative contributions from the different biomass burning regions (not shown). For example, SH fires contributed slightly more to the total CO in September-October 2011 (51–64%) than in 2012 (41–46%).

With respect to the domestic biomass burning sources, our analysis shows that tropical forest fires in the Amazon represents the largest source (52-75%) of the

total Amazonian biomass burning CO, followed by savanna fires (18-32%) and other fires, such as grassland and cropland (3-16%) (Figure 4.10c). As in the case of biomass burning CO from fire regions, we also find some interannual variability in the relative contribution from a specific biome (not shown). For instance, tropical forest fires in November contributed more to the total biomass burning CO in 2011 (72%) than in 2012 (68%). As mention previously, fire emissions in the Amazon are highly variable (Van der Werf et al., 2010), and specific weather conditions in a year i.e., spatial changes in drought location, can affect the type of biome burning (Gonzalez-Alonso et al., 2019) and hence, its relative contribution to total CO.

4.5.2 Contribution of biomass burning in the Amazon to CO and O_3

We compare CAMvert and CAMzeroBB simulations to assess the impact of Amazonian biomass burning on CO and O_3 across the Amazon domain. Figure 4.11 presents the relative difference in the simulated CO and O_3 column (0-8 km) in CAMvert for September 2012, and Figure B8 for March. Biomass burning in the Amazon contributes an average of 1% and 1.5% (March), and 8.5% and 5% (September) to the CO and O_3 background throughout most of the troposphere. However, similar to previous studies (Ziemke et al., 2009), locally, contributions can be larger. For instance, in September biomass burning in the Amazon contributed up to 58% and 30% $(17 \times 10^{17} \text{ molec/cm}^2 \text{ of CO} \text{ and } 9.2 \text{ DU of O}_3)$, respectively to CO and O_3 columns. Largest impacts are located where most CO and O_3 precursors are produced and transported downwind, across the arc of deforestation, along the Andes and towards the Pacific, in September. In addition, both simulations capture high tropospheric O_3 levels in the tropical south Atlantic, particularly in September $(\sim 30 \text{ DU})$. These are likely lower limit levels if we consider the low bias across the Atlantic mentioned in Section 4.4.2. High tropospheric O_3 levels have been previously reported over the South Atlantic during the burning season (Fishman et al.,

1996b, Edwards et al., 2003), but there is lack of consensus about its origin, i.e., biomass burning from the Amazon and Africa or lightning (e.g., Mauzerall et al., 1998, Thompson et al., 1996, Moxim and Levy, 2000, Sauvage et al., 2007, Ziemke et al., 2009). Our results show no contribution from Amazonian biomass burning to the high tropospheric O_3 levels over the South Atlantic in September.



Figure 4.11: Simulated averaged CO and O_3 columns by CAMvert and CAMzeroBB (a) and relative changes (in %; (\triangle Fire) between CAMvert and CAMzeroBB (b), in September 2012.

Consistent with previous studies (Kirchhoff et al., 1992, Galanter et al., 2000), Amazonian fires contribute to increasing surface O_3 mixing ratios at the source regions, up to 34% (6 ppb), in March and up to 87.5% (31 ppb), in September. Here, we characterise the impact of biomass burning in the Amazon on surface ozone levels that can be harmful to human health and vegetation. To quantify the impact, we use CAMvert and CAMzeroBB simulated hourly surface ozone mixing ratios and calculate metrics commonly used for human health, i.e., MDA8, and vegetation protection, i.e. M12 and AOT40, and compare them.

The maximum daily 8 h average ozone concentration (MDA8) is the metric most commonly used in many world regions for regulatory purposes concerning human health protection (e.g., Pfister et al., 2008, Reidmiller et al., 2009, Fleming et al., 2018). AOT40 is the sum of the difference between the hourly mean ozone value and values above 40 ppb for all daylight hours, considered here as from 0800 to 1959 hours (LT), over a specified period, usually the growing season (Mills et al., 2007). The daily 12-h (0800–1959 h) mean ozone exposure metric, M12 (e.g., Van Dingenen et al., 2009a, Avnery et al., 2011, Hollaway et al., 2012) has been widely used to characterize crop exposures to establish crop-specific exposure-response relationships. Figure 4.12 presents simulated CAMvert MDA8, M12 and AOT40, and fire-induced changes (\triangle Fire) i.e., difference with respect to CAMzeroBB during the burning season of 2012. CAMvert mean \pm standard deviation and maximum MDA8, M12 and AOT40 are 27.7 ± 6.99 ppb, 25.5 ± 6.63 ppb and 731 ± 1820 ppb h, and 53 ppb, 45 ppb and 15000 ppb h, respectively. Biomass burning in the Amazon contributes substantially to enhance these metrics. We observe maximum enhancements of 25 ppb, 20 ppb and 12000 ppb h, respectively for MDA8, M12 and AOT40, across the arc of deforestation, the Peruvian coast and Bolivia.



Figure 4.12: CAMvert simulated maximum daily 8 hour average (MDA8), daily 12-h (0800–1959 h) mean (M12) and sum of the difference between the hourly mean ozone (0800–1959 h) and values above 40 ppb (AOT40), and fire-induced changes (\triangle Fire, i.e., CAMvert-CAMzeroBB), during the 2012 burning season (July-November).

Impacts of fire-induced ozone on human health

Ozone standards for human health protection are variable across the globe. According to Fleming et al. (2018), MDA8 standards across the Amazon region range from 40–80 ppb. We set the MDA8 standard to 50 ppb in the domain and apply a limit of maximum 25 days of exceedance in a year. Figure 4.12 shows that the estimated MDA8 standard is exceeded across some regions in the SE Brazil and along the Pacific coast, during the biomass burning of 2012. However, some of these exceedances are likely to be produced by anthropogenic sources rather than biomass burning. We present in Figure 4.13 the estimated number of days with fire-induced exceedances of MDA8 O_3 during 2012. We observe that the fire-induced estimated MDA8 exceeds the standard in more than 25 days across regions where some of the most populated cities are located.



Figure 4.13: Number of days in which MDA8 O_3 was exceeded due to fire pollution (see text for more explanation). Main populated cities close to or within the excedances regions are also represented.

Figure 4.14 depicts the cumulative probability distributions of the estimated O_3 MDA8 during the burning season of 2012, over some populated cities within or close to fire-induced O_3 impacted regions. We also include estimated O_3 MDA8 from CAMsurf to show the impact of the injection height scheme for air quality purposes. Over most of these cities, we find a decline in air quality due to Amazonian fires, for most polluted days during the biomass burning season, with MDA8 values above 50 ppb. Furthermore, the estimated fire-induced impact is significantly reduced when the improved injection height scheme is applied (CAMvert), which highlights the necessity of an enhanced vertical distribution of biomass burning emissions in air quality studies. Previous studies showed that biomass burning is responsible for thousands of people's premature death across the world due to unhealthy air quality conditions (Jacobson, 2014). Furthermore, Reddington et al. (2015) estimated that aerosols from biomass burning in the Amazon can cause thousands of premature deaths. Here, we show that biomass burning in the Amazon contributes to increasing O_3 levels above the standards safe for human health, with the potential to affect more than 14 million people's health across the Amazon.

Impacts of fire-induced ozone on vegetation

Figure 4.12 shows that biomass burning is responsible for substantial enhancements in M12 and AOT40 during the biomass burning season of 2012. To establish vegetation-specific fire-induced impact that can be related to a reduction in production across the Amazon (Van Dingenen et al., 2009a, Mills et al., 2018), we calculate M12 and AOT40 for each crop-specific growing season or vegetation-specific period of maximum sensitivity. We focus on major crops for food security (i.e., wheat, rice), commercial crops which contribute significantly to the economy (i.e., soybean, sugarcane, cotton, coffee, tobacco, cocoa, banana and quinoa), and the tropical forest. First, we characterise the areas of fire-induced damage for each type of vegetation. We use datasets of crop harvested area and yield from 2000, on a 5 min by 5 min latitude and longitude grid (Monfreda et al., 2008). This dataset



Figure 4.14: Cumulative probability distributions of simulated surface O_3 MDA8 at the cities close or within regions with the highest number of days exceedances for CAMvert (blue), CAMsurf (red) and CAMzeroBB (black). City and population are shown in the insert.

comprises 175 distinct crops across the world, including our selection of crops. It should be noted that agriculture has largely changed (e.g. spatial extent) between the year of the Monfreda et al. (2008) dataset and the year under consideration. In the case of tropical forest, we use MODIS MCD12C1 Land Cover product (Friedl et al., 2010). We set the accumulation period of maximum sensitivity to ozone for each crop, i.e., the start and duration of the growing season. This is complex to determine as it depends on many factors, i.e. precipitation and temperature, which can be highly variable for a specific year or region. We extracted these data from crop calendars for South America, created by the United States Department of Agriculture USDA (USDA, 2019) and the Food and Agriculture Organization FAO (FAO, 2019), and crop-specific information¹. However, for some crops, these

¹https://www.fundesyram.info/biblioteca.php?id=4426; http://www.coffeeresearch. org/agriculture/environment.htm;http://www.agro20.com/; https://www.zipmec. com/en/south-america-season-for-fruits-and-vegetables.html; https://www.icco. org/faq/58-cocoa-harvesting/131-what-time-of-year-is-cocoa-harvested.html; http://www.soybeansandcorn.com/Brazil-Crop-Cycles; https://www.thebalance.com/

data are missing or given as national-level averages in graphical format. Therefore, we assume the specific-crop growing season for the main productive country and applied it to the harvested area within the domain. This assumption can be a source of bias, but in general, countries where the same crops grow are usually close in distance and tend to have similar ambient conditions, and thus growing season period. In reality, planting dates vary within each region and crop and, during the last years, production of many crops has increased due to double-cropping (Brazilian National Supply Agency, CONAB), which adds more complexity to determine each crop growing season. For tropical forest, we set the accumulation period during the biomass burning season.

Table 4.2 summarises the estimated CAMvert and CAMzeroBB M12 and AOT40 statistics for our selection of vegetation and crops, and Figure 4.15 shows the estimated fire-induced M12 and AOT40 for vegetation with a potential risk of yield loss. We observe that quinoa, tobacco, wheat and the tropical forest are highly sensitive to fire-induced O_3 , with maximum enhancements of the AOT40 of 9930 ppb h for quinoa and tobacco, 9510 ppb h for wheat and 12400 ppb h for the tropical forest. These enhancements are above the vegetation-based critical levels of 3000 ppb h during 3 months for wheat, and 5000 ppb h during 6 months for quinoa, tobacco and tropical forest, and can lead to production losses of 5–10% (Mills et al., 2018). Consistent with Pacifico et al. (2015), we find that the Amazonian forest productivity can be reduced by 5-10%, as the enhancement on AOT40 is more than double the critical level known to produce 5% loss. The growing season established for wheat, tobacco and quinoa are coincident with the biomass burning season. These crops are also produced across areas of Peru, Bolivia, Southern Brazil, where a large impact of fires on AOT40 and M12 is observed (Figure 4.12). Our results suggest that any crops with a growing season coincident with the biomass burning season and spatially distributed across these regions may be susceptible to yield loss due to high fire-induced O_3 from biomass burning in the Amazon. Furthermore, we

corn-planting-and-harvest-seasons-809309; http://esmiperu.blogspot.com/2007/08/ principales-cultivos-agrcolas-del-per.html; last access 14/02/2019

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4.2: Vegeta	etation with
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Crop	Growing season	Mean+SD	Maximum	$Mean\pm SD$	Maximum	Mean+SD	Maximum	Mean+SD	Maximum
Banana	Apr-Sep	1030 ± 1450	9410	$1640{\pm}1610$	10100	23.50 ± 8.09	39.0	$25.40{\pm}7.14$	38.9
\mathbf{Cocoa}	Nov-Apr	$1170{\pm}1840$	9580	$993{\pm}1730$	9740	$19.40 {\pm} 9.70$	43.2	20.30 ± 9.31	43.0
Coffee	Oct-Apr	$1570{\pm}2680$	25400	1440 ± 2510	25500	21.60 ± 9.43	42.7	$22.90{\pm}8.78$	42.6
Quinoa	May-Oct	1340 ± 2690	15600	3170 ± 3040	18500	16.70 ± 9.91	42.2	$23.80 {\pm} 9.06$	43.7
Sugarcane	Nov-Apr	1320 ± 2330	19400	1200 ± 2200	19000	$22.10{\pm}8.94$	43.2	$22.80{\pm}8.53$	43.0
Tobacco	May-Oct	$1340{\pm}2100$	15600	2250 ± 2620	18500	22.40 ± 9.22	42.2	$25.40{\pm}7.85$	43.7
Cotton	Dec-Feb	484 ± 988	10100	465 ± 963	9880	$23.80{\pm}7.98$	44.2	$24.30{\pm}7.66$	44.5
Maize	$\mathrm{Dec} ext{-feb}$	$604{\pm}1300$	10100	$554{\pm}1230$	9880	$22.10{\pm}8.64$	44.5	22.70 ± 8.32	44.5
Rice	$\mathrm{Dec-Feb}$	$680{\pm}1390$	10100	$614{\pm}1310$	9880	$21.90{\pm}8.87$	44.5	$22.50{\pm}8.54$	44.5
$\operatorname{Soybean}$	Jan-Apr	$911{\pm}1350$	12800	$899{\pm}1360$	12800	23.10 ± 8.25	40.7	23.50 ± 8.07	40.8
Wheat	Jul-Sep	$1090{\pm}1240$	10400	2220 ± 2060	13700	25.10 ± 9.56	45.6	29.70 ± 8.26	49.1
\mathbf{Forest}	Jul-Nov	$751{\pm}1540$	11400	1480 ± 1850	12400	$11.70{\pm}7.66$	38.9	$16.90{\pm}7.96$	39.0

attempt to estimate the relative yield loss (RYL) for wheat caused by fire-induced O_3 . We use Mills et al. (2007) concentration-response function and find a RYL of 5–23% for wheat, where regional fires may be responsible for 2-5% of the RYL. Most of the critical levels established by regulations are based on exposure-response studies for vegetation types and climate zones in the NH, mainly USA and Europe (Mills et al., 2007, 2018). Therefore, our results must be taken with caution. O_3 exposure-response functions specific to crops in the SH are necessary to allow for improved estimations on the O_3 impact and promote less sensitive O_3 crops across areas with a potential risk of exceedances of O_3 critical levels. We find that biomass burning in the Amazon can negatively impact the local economy through crops yield losses, which could be enhanced if fires increase, according to future projections.



Figure 4.15: Distribution of the tropical forest and fraction of harvested area for quinoa, wheat and tobacco in the Amazon domain (top) and simulated fire-induced surface O_3 AOT40 and M12 for quinoa, wheat, tobacco and tropical forest (bottom).

4.7 Conclusions

We implement a global smoke plume injection height parametrisation in CESM, that includes smoke plumes observed in the Amazon, to assess the performance on
the vertical distribution of CO and O_3 across the Amazon. We conduct two multiyear simulations: with emissions released at the surface and vertically distributed following the injection height scheme. Results from the simulations are compared with CO and O₃ observations from satellite, aircraft, ozonesondes, and ground-based measurements. Similar to previous studies (Jian and Fu, 2014, Zhu et al., 2018), the injection height scheme reduces CO and O_3 mixing ratios at the surface (70% and 40%), and enhances them in the free troposphere (30% and 10%) above the fires, especially during the peak of the burning season. On a regional scale, the injection height scheme produces overall CO column enhancement across downwind regions (5-10%) and reduction across the source regions (10-15%), which is consistent with the effect of injecting emissions at higher altitudes and the longer lifetime of CO in the FT, as argued previously (Chen et al., 2009). In the case of O_3 , an overall reduction in O_3 column is observed across the source regions (~5%), coincident with NO_x column reduction, the main limiting factor in O_3 production in the Amazon. The injection height scheme leads to improvements in the representation of CO and O_3 in the model, particularly near the source, with important regional air quality implications. However, we find some discrepancies with observations probably due to poor representation of biomass burning emissions and atmospheric processes in the model, which results in an excess of pollutants in the LT, mainly across the arc of deforestation and along the Andes.

We assess the relative importance of biomass burning in the Amazon with respect to other sources during 2012. As previously reported (Zeng et al., 2015), most of the CO in the Amazon is produced by oxidation of biogenic products (63%), given the relatively low anthropogenic emissions and the dominance of biogenic emissions. Biomass burning is the main source of directly emitted CO, from September to December (48–53%), comparable to anthropogenic emissions (43%) on an annual basis. Fires in the Amazon are the main source of CO from biomass burning from September to January (48-79%), although annually, long-range transport of plumes from Africa contributes significantly to fire CO (24–35%), as previously found (Edwards et al., 2006). Furthermore, most of the CO emissions from fires in the Amazon come from the tropical forest (52–75%) compared to other biomes. However, the contribution from biomass burning over a particular region or biome may change, as many factors are involved i.e., meteorological conditions or governmental policies (e.g., Reddington et al., 2015, Gonzalez-Alonso et al., 2019).

We conduct an additional simulation with fire emissions in the Amazon set to zero to assess the impact of domestic biomass burning on CO and O_3 during 2012. Consistent with previous studies (Galanter et al., 2000, Ziemke et al., 2009), biomass burning in the Amazon contributes significantly to enhancements of background CO (58%) and O_3 (30%) at the source regions. However, mean enhancements of CO are relatively small (8% in September) considering the large contribution from secondary production mentioned above. Fires in the Amazon are also responsible for important enhancements in surface O_3 (30 ppb) across the source and downwind regions, i.e., the arc of deforestation, along the Andes and towards the Pacific. We estimate the fire-induced O_3 impact on air quality with common metrics i.e., MDA8, M12 and AOT40. We find frequent exceedances of the O_3 standard for human health protection during 2012 and a decline in air quality over some of the most populated cities (1–2 million people) during the burning season. In addition, we find that fireinduced O_3 can lead to an estimated relative wheat yield loss of 2–5% and 5–10% of reduction in forest productivity, consistent with previous studies across the region (Pacifico et al., 2015). Moreover, any vegetation grown across the areas of major impact with a growing season coincident with the burning season may be at risk of production loss due to O_3 produced from biomass burning in the Amazon. Future projections suggest an increase in fires caused by climate change and anthropogenic activities, which may lead to higher ozone levels and degraded air quality across the region.

The results presented here show that biomass burning in the Amazon can considerably reduce O_3 air quality across the region, with negative consequences for health, ecosystems and local economy. Furthermore, we demonstrate the importance of an improved representation of the vertical distribution of biomass burning in models for future air quality studies. Ultimately, the outcomes of this study can help to efficiently manage air quality across the region and promote policies to limit fire burns.

Chapter 5

Summary and Conclusions

This work produced several major results. First, it characterised the vertical distribution of smoke plume heights from satellite observations across the Amazon and determined the main drivers that govern smoke plume dynamics across the region. Then, it demonstrated an improved representation of biomass burning products across the Amazon via implementation of a smoke injection height parametrisation in an Earth system model. Finally, it revealed that biomass burning in the Amazon substantially impacted surface ozone levels and degraded air quality across the region, with important risk for vegetation and population's health. The main conclusions of this work are presented below in more detail, followed by a general summary and recommendations for future research.

5.1 Vertical distribution of biomass burning emissions over the Amazon

Observations from MISR and MODIS (2005–2012) and CALIOP (2006–2012), allowed to derive an extensive climatology of smoke plumes across the Amazon, during the biomass burning season (July-November). The 8-year climatology showed larger number of smoke plumes during years with drought conditions, at the peak of the burning season (September), and over savanna and tropical forest, the dominant biomes in the region. The analysis on the vertical distribution of biomass burning emissions revealed that smoke plume heights exhibited large variability, from few hundred meters to 6 km, although most of the smoke was located below 2.5 km. In addition, smoke plume heights tended to be higher later in the afternoon, at CALIOP overpass, as a result of deeper PBL heights, possibly more energetic fires and CALIOP's greater sensitivity to very thin aerosol layers.

Finally, the Amazon climatology of smoke plumes demonstrated the value of combining observations from both instruments to constrain the vertical distribution of smoke from biomass burning over the Amazon.

5.2 Factors of variability on the vertical distribution of biomass burning over the Amazon

The 8-year climatology of smoke plumes across the Amazon combined with an extensive analysis on the main factors that drive smoke plume dynamics showed that smoke plume heights tended to be higher over biomes with dominant flaming combustion (i.e., grassland) compared to smouldering dominant biomes (i.e. tropical forest). Furthermore, a marked seasonal cycle on the smoke plume heights was noticed, where larger fire radiative power from the fire and more stable atmospheric conditions during the late burning season, increased the probability of fires to inject smoke in the FT, from 2–10%, in July to 15–40%, in November.

In addition, drought conditions increased fire activity and contributed significantly to the total aerosol loading across the region. Droughts were shown to favour understory fires and deeper PBL, where dominant smouldering combustion associated with low smoke injection heights, resulted in high smoke loading throughout the PBL, with important implications for regional air quality.

This work highlighted the importance of biome type, fire properties, atmospheric and, particularly drought conditions for plume dynamics and smoke loading in the Amazon.

5.3 Impacts of Amazonian biomass burning on surface ozone levels across the Amazon

The modelling experiments using the Community Earth System Model (CESM) with an updated smoke injection height scheme improved the representation of O_3 and CO in the model. The injection height scheme reduced O_3 and CO mixing ratios at the surface (40% and 70%) and enhanced them in the low-mid troposphere (10% and 30%) above the fires, which led to a better agreement with observations, particularly close to the source regions.

An analysis on the relative contribution from biomass burning to the total simulated CO across the Amazon domain during 2012 indicated that biomass burning was the main source of directly emitted CO from September to December, comparable to the contribution from anthropogenic sources on an annual basis. Furthermore, fires in the Amazon were the largest contributor to the total fire CO (36%) in 2012, although significant contribution from long-range transported plumes from Africa was evident (24–35%). Overall absolute contributions from biomass burning in the Amazon to total CO were considerably small (up to 8% in September) in the domain, where most of the CO was produced by oxidation of biogenic products (63%).

Results from the simulation showed that biomass burning in the Amazon was responsible for significant enhancements of surface ozone (30 ppb) in the peak burning season across the arc of deforestation, along the Andes and towards the Pacific, which could lead to unhealthy conditions. Particularly throughout 2012, estimated fire-induced O_3 produced frequent exceedances of the target value for the protection of human health (50 ppb) and induced a decline in air quality across some largely populated cities during the burning season. In addition, fire-induced ozone levels could also produce considerable damages in vegetation across areas of Peru, Bolivia and central and southern Brazil during the burning season. Furthermore, fire-induced ozone levels led to an estimated relative wheat yield loss of 2–5%, with implications for the local economy, and reductions in the tropical forest productivity of 5–10%.

This work provided important insights on the necessity of accurately represent the vertical distribution of biomass burning emissions for air quality studies and the impacts of biomass burning emissions in the Amazon on air quality.

5.4 Summary of conclusions and future research

The evidence presented in this work benefits future assessment on the impact of biomass burning emissions in the Amazon on air quality and climate. This work results in an improvement of the understanding of the vertical distribution of biomass burning pollution across the Amazon and the main factors of variability in plume dynamics. This information is crucial to predict and mitigate the impacts of biomass burning. In addition, this work provides evidence of the impacts of Amazonian biomass burning on surface O_3 levels during the burning season and the benefit of using an improved injection height scheme for air quality studies. The results and analysis exhibited in this work motivate further investigation. Some opportunities for future research are presented below.

First, the climatology of smoke plume heights across the Amazon established a relationship between smoke plume heights, smoke loading and drought conditions. In view of more frequent droughts expected in the near future, an extended analysis of smoke plumes including 2015, associated to severe drought conditions induced by el Niño, is recommended to establish trends on plume dynamics across the region and understand the impact of increased frequency of droughts.

Results from the simulation experiments evidenced an excess of biomass burning pollution within the boundary layer across the Amazon. To understand the role of an overestimation of biomass burning emissions or a poor representation of atmospheric processes in the model i.e., convection, a simulation with different biomass burning emission inventories, such as GFED, will help shed light on this discrepancy.

Appendix A

Supplementary information

This document includes supplementary tables, figures and results used for the analysis discussed in Chapter 3 of this work.

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ıble
A1:
Statistical
summary
for
main
smoke
plume :
and
plume
rise
controlling
$parameters^a$.

		Tropica	1 Forest			Sava	nna			Grass	land		
	Dry Y	lears ^b	Wet Y	lears ^b	Dry	Years	Wet 1	Years	Dry Y	lears	Wet Y	Years	
	$Early^{c}$	$Late^{c}$	Early	Late	Early	Late	Early	Late	Early	Late	Early	Late	
Smoke Height (m)	698 ± 407	898 ± 451	906 ± 418	1180 ± 693	795 ± 462	1110 ± 589	984 ± 473	1250 ± 653	881 ± 508	1160 ± 474	1280 ± 971	1350 ± 728	
FRP(MW)	182 ± 408	245 ± 613	369 ± 813	369 ± 813	352 ± 744	503 ± 828	269 ± 456	330 ± 537	390 ± 480	567 ± 838	148 ± 152	529 ± 654	
AOD (unitless)	0.53 ± 0.29	0.73 ± 0.37	0.21 ± 0.17	0.33 ± 0.21	0.37 ± 0.30	0.46 ± 0.32	0.14 ± 0.12	0.28 ± 0.18	0.29 ± 0.26	0.58 ± 0.35	0.1 ± 0.07	0.30 ± 0.15	
Stability (K/km)	5.27 ± 3.64	4.28 ± 2.82	3.93 ± 2.17	3.11 ± 1.76	4.17 ± 3.72	2.92 ± 2.95	3.44 ± 3.24	2.47 ± 2.26	2.45 ± 3.07	2.32 ± 1.92	3.15 ± 3.65	2.07 ± 1.53	
PBL Height (m)	1330 ± 535	1290 ± 551	1150 ± 487	1220 ± 486	1580 ± 509	1510 ± 482	1350 ± 522	1480 ± 543	1760 ± 525	1770 ± 534	1360 ± 443	1380 ± 316	
Number	536	438	238	271	434	575	353	516	51	43	17	38	
^a Reported the averag	e±SD for smok	e plumes with v	alid values for	all the paramet	ers								

^aReported the average±SD for smoke plumes with valid values for all the parameters ^bDry years are 2005, 2007 and 2010 and wet years are 2006, 2008, 2009, 2011 and 2012

 $^c\mathrm{Early}$ season is defined as July–August and late season as September–November



Figure A1: Percentage of MISR plumes in the climatology classified by year, month, biome and drought conditions. Absolute values in each distribution are included above the bars. Percentage of MISR plumes classified by biome and year are also shown, as tropical forest (green), savanna (red), grassland (blue), and cropland and not classified (grey).



Figure A2: Vertical distribution of individual MISR stereo-height retrievals, averaged over all plumes in the eight-year of the study and stratified by biome.



Figure A3: Relationship between MISR maximum plume heights and MODIS total fire radiative power (FRP) for the 8-year data set. Data are colour-coded per biome as tropical forest (green), savanna (red), grassland (blue), and cropland (grey). Total number of observations and r^2 are given in the annotation of each panel.



Figure A4: Interannual variability of MISR plume maximum heights above the terrain, MODIS FRP and MISR AOD by biome. Bar plots indicate the distribution of the data for each year. Bars in this figure are ordered based on DSI (Table 3) rather than chronologically, The medians (grey circles) and the means (black squares) are shown along with the central 67% (box) and the central 90% (thin black lines). Distributions are colour-coded based on drought conditions (Table 3). Note that, although no MODIS DSI data are available for year 2012, this year is plotted in the middle as it is defined as dry year by Erfanian et al., (2017). The number of observations (in black) and the median values (in red) included in each distribution are given at the top of the plot.



Figure A5: Location of the CALIOP plumes analysed with the median smoke plume height over the Amazon domain.



Figure A6: Examples of CALIOP vertical extinction profiles with smoke aerosols above 6 km. Values are coloured by classified aerosol types. Profile 20070923D represents a case where high altitude smoke is disconnected from low altitude smoke and profile 20100831N gives an example of smoke gradually distributed from low to high altitudes.

Appendix B

Supplementary information

This document includes supplementary tables, figures and results used for the analysis discussed in Chapter 4 of this work.



Figure B1: Fire regions of tagged CO across the globe used. EURO: Europe, BONA: Boreal North America, BOAS: Boreal Asia, TENA: Temperate North America, CEAS: Central East-Asia, SEAS: South East-Asia, EQAS: Equatoria-Asia, INDI: India, AUST: Australia, SAM: South America, CEAM: Central America, NHAF: North Africa, SHAF: South Africa, AMAZ: Amazon.

Table B1:Summary of the	TOAR stations for	r surface ozone eva	luation.
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Name	Station type	Area type	Country	Longitude	Latitude	Altitude
Porto Velho	background	rural	Brazil	-63.86	-8.68	84
Ragged Point	other-marine	rural	Barbados	-59.43	13.16	15
Amazon TT34	background	remote	Brazil	-60.209	-2.594	150
Cayenne	background	urban	French Guiana	-52.33	4.93	4
Cape Verde	background	remote	Cape Verde	-24.87	16.84	10
San Lorenzo	background	remote	Paraguay	-57.55	-25.36	133
Tololo	background	remote	Chile	-30.17	-70.79	2220



Figure B2: Location of the ozone observational dataset used in our model evaluation. TOAR stations with surface ozone measurements (orange square), ozonesondes in the Ecuatorial region (blue square) and ozonesondes in the Atlantic/African region (red upside down triangle).



Figure B3: MODIS active fires and FINNv1.5 CO and NO $_x$ emissions in March and September 2012.



Figure B4: CAMsurf and CAMvert simulated CO mixing ratios (ppb) at three different levels: the surface, 2 km and 6 km (a), and the column (0–8 km) (molec/cm²) for March and September 2012.



Figure B5: Averaged MOPITT CO mixing ratios and relative bias (in %) of CAMsurf and CAMvert with respect to MOPITT at 400 and 800 hPa for March 2012.



Figure B6: CAMsurf and CAMvert simulated O_3 mixing ratios (ppb) at three different levels: the surface, 2 km and 6 km (a), and the column (0–8 km) (DU) for March and September 2012.



Figure B7: Monthly averages of O_3 mixing ratios from TOAR observations (black), CAM-vert (blue), CAMsurf (red) and CAMzeroBB (green). Vertical bars indicate the standard deviation of the mean.



Figure B8: Simulated averaged CO and O₃ columns by CAMvert and CAMzeroBB (a) and relative changes (in %; (\triangle Fire) between CAMvert and CAMzeroBB (b), in March 2012.

Appendix C

Contributions and co-authors

Chapter	Contribution
1	My own work.
2	My own work.
3	This chapter was published on February 8th, 2019 in the Atmospheric
	Chemistry and Physics journal (ACP) with the following co-authors:
	Maria Val Martin and Ralph A. Kahn.
	I carried out the study, developed the climatology, analysed the data,
	and drafted the manuscript. Maria Val Martin was the principal in-
	vestigator of the study, and supervised my analysis and manuscript
	writing. Ralph A. Kahn contributed to the interpretation of results and
	assisted on writing.
4	This chapter is prepared to be submitted with some modifications to
	ACP journal with the following co-authors: Maria Val Martin, Merritt
	N. Deeter, Benjamin Gaubert, Louisa K. Emmons and Simone Tilmes.
	I carried out the study, prepared the modelling experiments, analysed
	the results and drafted the manuscript. Maria Val Martin gave me ad-
	vice and guidance on the study, and supervised my analysis and manu-
	script writing. Merritt N. Deeter provided MOPITT data and expertise
	guidance on MOPITT products. Benjamin Gaubert provided expertise
	guidance on CESM and helped me to prepare the modelling experi-
	ments and the CO tag experiment. Louisa K. Emmons provided the
	emission files at CESM grid and expertise guidance on CESM. Simone
	Tilmes provided the MERRA-2 meteorological fields at CESM grid, the
	ozonesondes dataset and expertise guidance on CESM.
5	My own work.

Table C1: Summary of contributions and co-authors.

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