

The vertical distribution of biomass burning pollution over tropical South America from aircraft in situ measurements during SAMBBA

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15 **Abstract.**

~~To reduce the uncertainties in processes driving the vertical distribution of biomass burning pollutants, and thus impacts on regional weather and climate, we present an integrated analysis of vertical profiles of pollutants and meteorological parameters measured on flights during the 2012 South American Biomass Burning Analysis (SAMBBA) field experiment. We examine processes driving the vertical distribution of biomass burning pollution following an integrated analysis of over 200 pollutant and meteorological profiles measured in-situ during the South American Biomass Burning Analysis (SAMBBA) field experiment. This study will aid future work examining the impact of biomass burning on weather, climate and air quality.~~

During the dry season there were significant contrasts in the composition and vertical distribution of haze between western and eastern regions of tropical South America. Owing to an active or residual convective mixing layer, the aerosol ~~burden~~ abundance was similar from the surface to ~1.5 km in the west and ~3 km in the east. Black carbon mass loadings were double
25 in the east ($1.7 \mu\text{g e m}^{-3}$) than west ($0.85 \mu\text{g e m}^{-3}$) but aerosol scattering coefficients at 550 nm were similar ($\sim 120 \text{ Mm}^{-1}$), as
too were CO near surface concentrations ($310\text{-}340 \text{ ppb}$). We attribute these contrasts to the more flaming combustion of Cerrado fires in the east and more smouldering combustion of deforestation and pasture fires in the west. Horizontal wind shear was important in inhibiting mixed layer growth and plume rise, in addition to advecting pollutants from the Cerrado regions into the remote tropical forest of central Amazonia. ~~Optically thin~~ Thin layers above the mixing layer indicates roles
30 for both plume injection and shallow moist convection in delivering pollution to the lower free troposphere. However, detrainment of large smoke plumes into the upper free troposphere was very infrequently observed. Our results reiterate that thermodynamics control the pollutant vertical distribution and thus point to the need for correct model representation so the spatial distribution and vertical structure of biomass burning smoke is captured.

~~We observed an increase of aerosol abundance relative to CO with altitude both in the background haze and plume enhancement ratios. It is unlikely associated with thermodynamic partitioning, surface aerosol deposition or from non-biomass burning sources and we speculate it may be linked to fire combustion efficiency coupled to plume injection height. Further enquiry is required to explain the phenomenon and explore the impacts for regional climate and air quality.~~

5 ~~— Our observations of relatively large concentrations of aerosol aloft and of CO near surface suggest that there is a greater contribution of pollutants from more complete combustion with altitude. Release of appropriate emissions from the initial more flaming and later residual smouldering phases of a fire at appropriate altitudes may be especially important to ensure models can accurately predict aerosol radiation, aerosol cloud and air quality impacts.~~

1 Introduction

10 The vertical distribution of biomass burning pollution influences its impacts on weather (e.g. Kolusu et al., 2015), climate (e.g. Boucher et al., 2013), human health (e.g. Reddington et al., 2015) and ecosystem function (e.g. Rap et al., 2015). The vertical location of aerosol is particularly sensitive since this can impact radiative forcing (Zarzycki and Bond, 2010), alter cloud microphysics (Tao et al., 2012) and feedbacks on cloud dynamics (Feingold et al., 2005). The altitude of pollutants also determines their atmospheric residence time, which will affect any ageing processes and the resultant horizontal distribution
15 following advection. Model predictions of the vertical distribution of the effects of biomass burning smoke are therefore very dependent on accurately capturing the vertical structure, especially in regions of the tropics where land use change is rapid and smoke is widely distributed. However, a lack of detailed observations across these regions limits model constraint.

These uncertainties are pronounced in tropical South America (TSA), one of the largest global biomass burning sources. Aerosol accumulates within the convective boundary layer forming a regional haze ~~with a lifetime of 4-5 days (Edwards et al.,~~
20 ~~2006)~~ that can cover up to 6 million km² (Prins et al., 1998) ~~and whilst weekly averaged aerosol optical depths (AOD) are typically 0.75-1 in the mid visible, they can reach 4 in extremely polluted years associated with drought and reach aerosol optical depths (AOD) of up to 4 in the mid visible during particularly polluted years~~ (Artaxo et al., 2013). Fires are widespread within the so-called ‘arc of deforestation’ which traverses the southern and eastern edges of the Amazon basin. This arc comprises both tropical forest rainforest and Cerrado (savannah-like) biomes which feature distinct fire regimes.

25 Previous studies have discussed the vertical distribution of biomass burning pollutants in TSA based on in situ aircraft measurements. The 1985 ABLE-2B study across northern Amazonia from the coast to Manaus provided profiles and transects of aerosol extinction (Andreae et al., 1988), CO (Sachse et al., 1988), aerosol number and ozone (Gregory et al., 1988). The 1992 TRACE-A campaign in central and eastern TSA yielded vertical profiles of total aerosol and black carbon mass (Pereira et al., 1996). The 1995 SCAR-B and 2002 LBA-SMOCC studies focused on western TSA, providing vertical information on
30 aerosol number (Andreae et al., 2004), aerosol scattering (Chand et al., 2006; Ross et al., 1998) and CO concentrations (Freitas et al., 2009). The 2004 TROFFEE campaign in central TSA provided vertical profiles of CO and CO₂ (Yokelson et al., 2007). Finally, the 2009 BARCA-A study provided aerosol number and CO concentrations in central and northern TSA (Andreae et

al., 2012). Common among these studies is the limited regional spatial extent and the coverage above ~4 km, whilst many only show singular profiles and/or do not consider the eastern Cerrado fire regime. Furthermore, profiles are typically not integrated with multiple pollutants nor simultaneous thermodynamic measurements. This is important for understanding how mixing, advection and removal affect pollutants differently. Likewise, recent lidar remote sensing from satellite (Bourgeois et al., 2015; Huang et al., 2015) and surface (Baars et al., 2012) platforms do not provide the comprehensive dataset best suited to examining the drivers of multi-pollutant vertical distribution.

When simulating the pollutant vertical distribution over TSA there is a large diversity between AeroCom global models (Koffi et al., 2012) and between a subset of regional models (Andreae et al., 2012). In particular, open questions remain regarding treatment of convective mixing (Archer-Nicholls et al., 2015) and the injection of buoyant plumes into the vertical column (Paugam et al., 2016). Furthermore, to ensure consistency with remotely sensed AOD measurements, aerosol emissions are typically up-scaled by a factor of two to five (Johnson et al., 2016; Reddington et al., 2016a). ~~There is no established physical understanding of this requirement, with~~ This represents a key uncertainty in model treatment of biomass burning aerosol, with deficiencies in both emissions inventories and model process posited. ~~As such, scaling factors represent a key uncertainty in model treatment of biomass burning across the globe.~~

In this study we examine the key controls on the vertical distribution of biomass burning pollution across TSA during the 2012 dry season using data collected on research flights during the SAMBBA (South AMerican Biomass Burning Analysis) field experiment. This study includes the first incandescence based measurements of black carbon mass and mixing state in a tropical biomass burning region, and also the most comprehensive integrated multi-parameter library of profiles for tropical South America. It also represents the first comprehensive airborne sampling in eastern Cerrado regions of intensive fire activity. To determine the key drivers of pollutant distribution we examine: features of the atmospheric structure (Sect. 3.1); their interaction with pollutants based on a novel and detailed analysis of individual profiles (Sect. 3.2); and, fire activity and horizontal aerosol distributions based on satellite observations (Sect. 3.3). Average pollutant profiles are presented across the different synoptic regimes over TSA (Sect. 3.4). Together, these approaches allow determination of the key drivers of the vertical distribution of biomass burning pollution, and an examination of how these manifest in different environments and relate to previous observations (Sect. 4). A discussion of implications and recommendations for future measurement and model approaches to reduce the uncertainty surrounding the impacts of biomass burning pollution both in TSA and other tropical biomass burning regions is presented (Sect. 5).

2 Experimental details

Twenty science flights were conducted during the SAMBBA experiment between 14th September and 3rd October 2012, primarily based out of Porto Velho in northern Rondônia state, Brazil (Fig. 1). Extensive geographical sampling was conducted via three basic flight operations: (i) straight and level runs within the boundary layer, (ii) high altitude surveys and (iii) profile ascents/descents. Flights were conducted by the UK BAe-146 research aircraft operated by FAAM (Facility for Airborne

Atmospheric Measurements). The BAe-146 flew with a comprehensive instrumentation suite, capable of measuring aerosols, dynamics, cloud physics, chemical tracers and meteorological fields (McBeath, 2014). Simultaneously, a fully instrumented ground site was operational in Porto Velho, the results from which are described in Brito et al. (2014).

Aerosol total scattering coefficients were measured by a 3-wavelength integrating nephelometer (TSI Inc., USA; Anderson et al., 1996). Standard corrections were applied for angular truncation and non-Lambertian light source errors (Anderson and Ogren, 1998; Müller et al., 2011). The total scattering coefficient at 550 nm is reported at either dry RH (<30%), $\sigma_{sp,dry}$, or ambient RH, $\sigma_{sp,amb}$. These values were derived from the measured data based on the internal RH of the nephelometer and applying humidification scaling factors from Fig. 4c of Kotchenruther and Hobbs (1998). Note, however, there remains significant uncertainty on the role of water uptake on aerosol in TSA (Reddington et al., 2018, Darbyshire et al., in preparation).

Mass concentrations of refractory Black Carbon (rBC) were obtained using the Single Particle Soot Photometer (SP2, Droplet Measurement Technologies, USA; Baumgardner et al., (2004), Stephens et al., (2003)). The operation on board the BAe-146 is outlined by McMeeking et al. (2010) and the calibrations and post processing undertaken for this work are described by Allan et al. (2014). Reported mass loadings have a measurement uncertainty of approximately 30% (Schwarz et al., 2008; Shiraiwa et al., 2008). The coating thickness of scattering material on core rBC particles was calculated following the method presented by Liu et al. (2014) and Taylor et al. (2015). By assuming a full concentric encapsulation of the spherical core, the coating thickness of single rBC particles are estimated using a core refractive index of 2.26-1.26i and coating refractive index of 1.50+0i (Taylor et al., 2015).

The SP2 and nephelometer sampled through a Rosemount inlet which has a high transmission for the submicron aerosols ~~presented-of interest~~ here (Trembath, 2013). rBC and σ_{sp} are reported at standard temperature (273.15 K) and pressure (1013.2 hPa) to allow direct comparison of particle composition at different altitudes.

Carbon dioxide mixing ratios were measured by a fast greenhouse gas analyser (FGGA; Los Gatos Research Inc.) adapted for aircraft use (O'Shea et al., 2013). The total accuracy of this measurement is estimated at ± 0.17 ppm. Carbon monoxide mixing ratios were measured by an Aero-Laser AL5002 VUV resonance fluorescence gas analyser (Gerbig et al., 1999). Total uncertainty of the FAAM instrument is estimated to be $\pm 2\%$ (O'Shea et al., 2013). In flight calibrations were performed for CO and CO₂ using World Meteorological Organization traceable gas standards.

A General Eastern 1011B (GE Measurement & Control) chilled mirror hygrometer provided measurements of ambient dew point temperature, accurate to ± 0.2 °C. A Rosemount/Goodrich type-102 True Air Temperature sensor was mounted outside the aircraft, providing ambient temperature measurements using a Rosemount 102AL platinum resistance immersion thermometer. ~~A five hole radome-mounted turbulence probe at the aircraft nose provides measurement of airflow relative to the aircraft, thus allowing calculation of wind vectors when combined with a GPS inertial navigation unit~~ (Petersen and Renfrew, 2009). A total of 56 Vaisala RD94 dropsondes were released from the aircraft at altitudes up to 8 km, providing geolocated profiles of pressure, temperature, relative humidity and wind vectors to ground level.

Individual profiles from the aircraft and dropsondes were averaged (median) onto a 50 m vertical grid for automated identification of thermodynamic features and their relation to haze distribution and pollutant plumes. Plumes identified on take-off/approach to an airport were removed from further analyses as they were likely contaminated from urban emissions.

Satellite remote sensing products were obtained for the SAMBBA period and September 2008-2017. The MODIS (Moderate Resolution Imaging Spectroradiometer) instrument provided retrievals of aerosol optical depth and active fire data. Precipitation rates were acquired from instruments on-board TRMM (Tropical Rainfall Measuring Missions). Land cover data was acquired from the ESA CCI Land Cover project and used the United Nations Land Cover Classification System. Specific details on the acquisition and processing of satellite data are provided in the supplement.

3 Results

10 3.1 Atmospheric structure

The evolution of the convective boundary layer is important for pollutant mixing, advection and plume injection and is idealised in Fig. 2. Surface heating initiates buoyant turbulent motions which deepen throughout the day and comprise the convective mixing layer. In the morning these thermals slowly destabilise the nocturnal stable layer before rapidly penetrating into the residual layer, i.e. the previous days' boundary layers. Growth of the mixed layer is inhibited by the statically stable entrainment zone, a region of overshooting thermals and downward entrainment from above. The lifting condensation level is typically within the entrainment zone but above the mixed layer top, leading to patchy cumulus fields. These clouds are either forced, i.e. fair weather cumulus which do not vent mixing layer air into the free troposphere, or active, where the level of free convection is reached and mixing layer air is vented through the tops of towering cumulus that may grow up to the limit of convection and detrain in the upper troposphere (Stull, 1985). Typically, a maximum in horizontal wind speed of variable magnitude and extent is present above the entrainment zone and referred to as the trade wind inversion.

The layer altitudes in Fig. 2 are based on a semi-automated analysis of individual thermodynamic profiles, the methodology for which is detailed in Sect. S2. This analysis is summarised for an example profile in Fig. 3A-C. From the wind speed and direction profiles (panel A), regions of local positive wind shear (vertical brown line) and locations of wind speed maxima (open purple circles) are identified. Profiles of relative humidity, specific humidity, potential temperature and equivalent potential temperature (panel B) act as the basis for identification of a mixed layer and where so, its depth. This involved a manual approach based on the spread of a number of automated methods. Profiles of temperature and dew point temperature (panel C) are presented in a skew-T log-P format, allowing calculation of the lifting condensation level (LCL), level of free convection (LFC) and limit of convection (LOC). The entrainment zone was not derived due to difficulties in determining its base with the information available. The format of Fig. 3 is replicated for each profile and displayed in Sect. S5.

30 In general, observed profiles of horizontal wind speed reflect those expected based on our understanding of synoptic flows over TSA_(Campetella and Vera, 2002). In the lower troposphere (850 hPa) the mean synoptic circulation is an anticyclonic flow centred around south eastern TSA: easterly trade winds turn south-eastward toward the extra tropics, parallel to the Andes

(Fig. 4C). Horizontal wind speed is greatest along eastern coastal regions, northern Amazonia and Bolivia, whilst slacker wind speeds are prevalent in south western TSA. At higher levels the influence of the Andes is reduced, resulting in a more zonal flow.

5 There is considerable structure in the observed profiles which is not captured by large scale wind fields, such as those in the ECMWF ERA-Interim reanalysis. This includes multiple filament like maxima in wind speed, often without accompanying changes in wind direction. In western TSA the expected sharp change in wind direction and increase in horizontal wind shear associated with a trade wind inversion is often not present and instead a more gradual change in speed and direction was observed. In the mixed layer, horizontal wind speed is typically lower and of a more uniform direction and magnitude. There are a number of exceptions to this generalisation, where positive wind shear regions or wind speed maxima are present through
10 the mixed layer. A wind speed maximum is generally observed above or collocated with the mixed layer top (e.g. Fig. 3). At times this was the trade wind inversion but typically it was not. If this feature is associated with a region of positive wind shear below, mixed layer growth is likely inhibited and in turn the magnitude of the jet and gradient of wind shear reduces.

Variations of the Fig. 2 idealised structure were also found to depend on the sample region, owing to the west-east asymmetries in land use. Compared to the Cerrado regions, the northern and western tropical forest regions experience higher
15 levels of precipitation, soil moisture and cloud fraction resulting in lower solar insolation and surface temperatures (Fig. S2). On average these resulted in a deeper, faster developing, mixing layer in the east than west and north (average of 1.8 km vs. 1.1 km). As such the lifting condensation level (2.6 km vs. 1.4 km) and level of free convection (3.1 km vs 2.4 km) was also greater. Only in 25% of profiles was the limit of convection identified, where it was higher in the east than west and north (4.4 km vs. 3.5 km).

20 Superimposed on the regional variation was the impact of two distinct meteorological phases in north and west TSA. Between 14-Sept and 22-Sept conditions were characteristic of the dry season, whilst between 23-Sept and 05-Oct conditions more closely resembled those of monsoonal transition (Brito et al., 2014). This second phase was characterised by an increased rate and area of rainfall, accompanied by reductions in solar insolation and increases in cloud fraction and soil moisture (Fig. 4). In ~30% of profiles no mixing layer could be identified and instead the lower atmosphere was conditionally unstable
25 throughout (and hence Fig. 2 is unrepresentative). This typically occurred in the west of TSA and toward the end of the campaign as wet season type conditions became more prevalent. Meteorological conditions during the 2012 SAMBBA period were typical of the climatology – there were no climatic extremes (Fig. S2).

3.2 Determining pollutant vertical distribution via feature analysis of individual vertical profiles

An automated analysis of interactions between atmospheric structure and pollutant burdens-abundance was undertaken for
30 each individual profile. Results from this analysis are summarised in Table S1 and illustrated for the example profile in Fig. 3D-G. The full analysis methodology is provided in Sect. S2.

When present, the convective mixing layer was investigated to determine if each pollutant was correspondingly well mixed throughout the layer. This is the case in approximately half of all profiles featuring a mixed layer. The profiles where this was

not the case are typically perturbed by fresh plumes and/or wind shear and jet interactions. The number of CO and CO₂ profiles taken during the morning that were classified as well mixed was approximately half that of the afternoon. For example in Fig. 3 - as indicated by the vertical green line and accompanying yes/no indicator - rBC and σ_{sp_dry} are considered well mixed but CO and CO₂ are not, due to concentrations increasing near the surface.

5 If pollutant ~~burdens-abundance~~ in the region above the mixing layer (or if absent, the near surface) and 4 km exceeded those of unpolluted background conditions, a pollutant residual layer was identified. Dry season background conditions were defined as 0.1 $\mu\text{g em}^{-3}$ for rBC (Artaxo et al., 2013), 15 Mm^{-1} for σ_{sp} (Rizzo et al., 2013) and 140 ppb for CO (Andreae et al., 2012). Since CO₂ has significant global trends and the large biogenic sources/sinks its background is harder to quantify and is not considered here. Over ~~8970%~~ of profiles included a pollutant residual layer of rBC, σ_{sp} and CO, even those in remote
10 regions away from fresh emissions (Table S1). Typically, the shape of these residual layers is similar to that from the previous day(s) boundary layer. However there is substantial diversity in shape and magnitude, especially between 2 - 4 km that result from variations in plume injection, long range transport, cloud detrainment or entrainment of clean air from aloft. This is seen in the profiles in Fig. 3 where the residual layer is indicated by the vertical brown line and accompanying yes/no indicator.

Two impacts of horizontal wind shear on pollutant vertical structure were explored. Firstly capping effects, indicated by a
15 decline in pollutant abundance burdens over regions of positive horizontal wind shear. This was present in 40-60% of instances. Secondly the curtailment by wind shear on plume rise via increased drag and lateral entrainment, as previously explored by Freitas et al. (2010). This was indicated by a co-location of pollutant abundance burden-maxima with wind shear maxima. This was present in 20-30% of instances. Fig. 3 contains such an example at ~650 hPa, as illustrated by the purple circle in panel F.

20 Plumes were identified by a significant enhancement in pollutant abundance burden (dot-dash grey lines, Fig. 3.D-G) over a local background (dashed grey lines, Fig. 3.D-G). The majority of profiles contain a plume of one or more pollutant. Only in 10% of these plumes did all pollutant abundances burdens exceed the identification threshold, despite often being well correlated, indicating variability in plume composition. The plumes observed in the mixing layer are likely more fresh and locally injected. Their frequency increases in the afternoon (Table S2) which may be associated with increased fire incidence
25 (Giglio et al., 2006). Plumes were encountered between the mixing layer and lifted-lifting condensation level in 15-20% of profiles, indicating a significant number of fire plumes have sufficient energy to overcome the stable entrainment zone. If the pyro-convection is such that the plume reaches above the lifted-lifting condensation level, as in 10-25% of profiles, pyro-cumulus form and detrain pollutants. Plumes encountered at these levels likely include those from local sources and those advected from regional upwind sources. The frequency of these plumes generally declines in the afternoon indicating pollutants
30 have been re-entrained into the mixing layer or, in western regions, transported following deep moist convection above our flight ceiling.

It is possible the layers of enhanced pollutants above the lifting condensation level are not direct plume injections but detrainment from active cumuli following shallow moist convection. However, mixing layer pollution transported in this manner will be of similar concentrations to that of the residual layer and as such may not be significant enough to trigger the

plume thresholds defined here. Furthermore it would be expected that relative concentrations of CO would be conserved but those of aerosol would be reduced following activation and wet scavenging if precipitation occurred. However, analysis of the plume composition (Sect. 3.4) indicates the opposite to be true. Together this indicates the pollutant enhancements observed ~~here via this analysis~~ are likely plumes rather than detrained layers. There is evidence of moist convection delivering CO to altitudes above ~4 km but with significant wet scavenging of aerosol. In 81% of profiles with sufficient vertical coverage CO loadings increased by 40 ppb from a minimum at ~4km to the top of the profile. Unlike the discrete signal from plumes the enhancement was often 1-2 km deep. Of the rBC and σ_{sp} profiles only 8% and 3% had a similar increase in signal co-incident with CO enhancements. This indicates significant removal of aerosol from deeper convection to altitudes above ~4km, although moist shallow convection is still likely important in transporting mixing layer air aloft. In the subset of profiles where the limit of convection was within the flight envelope only a couple of plumes were identified.

3.3 Fire activity

During the dry season fires are prevalent across TSA but especially between 0 and 25°S (Fig. 5A). Pollutants emitted from these fires accumulate in south-western TSA as the Andes act as a barrier to smoke advected westward via the trade winds (Fig. 5C). This regional smoke plume is optically thick and its shape and magnitude are dependent on fire activity in a given year. Whilst the September 2012 dry season is generally representative of years since 2008 (Fig. 5A-C ii.), fire count and radiative power were greater in the eastern states. This likely explains ~~the enhanced~~ an enhancement in AOD of ~0.1 in these regions and southern Mato Grosso state. The shift to wet season conditions corresponded to a reduction in fire count (Fig. 6A) and emissions (Pereira et al., 2016). We speculate conditions were not optimal for human ignited fires, as a rise in relative humidity and increase in wind speed makes ignition and control more difficult, whilst decreasing the fuel consumption. The increase in cloud cover may have also reduced fire detection efficiency. In northern, western and central regions we speculate this is likely due to a reduced ignition probability and the dampening or extinction of existing burns. The reduction in fire count and intensity in eastern regions occurred despite no significant change in precipitation. We speculate this may result from stronger easterlies (Fig. 4C.iii) inhibiting fire ignition and spread. Together with increased wet removal, these effects acted to decrease the regional AOD by ~0.2 and shift the peak AOD from central to north eastern regions (Fig. 6C).

3.4 Vertical distribution of pollution over synoptic scales

Based on the variability between the individual profiles and the regional differences in fire activity, land use and meteorology, we define four regimes which characterise the key synoptic differences in pollutant ~~abundance burden~~ and vertical distribution across TSA during SAMBBA:

- Deforestation impacted western Amazonia around Rondônia and Mato Grosso states (68.5°W - 54°W, 12°S - 5.5°S) during dry season conditions (14-Sept to 22-Sept). This regime is henceforth referred to as W1.
- The same area during dry-wet transition season conditions (23-Sept to 05-Oct). Henceforth W2.

- ‘Pristine’ rainforested area north of Manaus (65°W - 59°W, 5.5°S - 1°S) during the dry season on 19th Sept. Henceforth N1.
- The dry Cerrado environment in eastern regions around Tocantins state (53°W - 46°W, 12°S - 9°S) between 26th Sept. and 2nd Oct. Henceforth E0.

5 The geographic extent of these regimes is demonstrated in Fig. 1 and the median thermodynamic variables in Fig. 7.

The average aerosol vertical profile shapes of W1, W2 and N1 are similar (Fig. 8.A,B) - similar loadings occur from the near surface to 3 km with a small maximum between 1 and 2 km. Above 4 km, loadings decline to near background values. Comparatively, E0 is characterised by a near surface maximum, which may be because multiple fresh fires were sampled, followed by similar loadings up to 4 km, above which concentrations declined sharply to near zero. The shape of the aerosol profiles is not significantly affected if plumes are removed from the analysis.

There are also contrasts in the ~~absolute~~-aerosol abundance burden between the regimes. There is a twofold reduction throughout the column between W1 and W2, consistent with the satellite AOD fields (Fig. 6). Within the boundary layer this represents a reduction from 0.85 to 0.4 $\mu\text{g em}^{-3}$ for rBC and 115 to 60 Mm^{-1} for $\sigma_{\text{sp,dry}}$. rBC loadings of 0.85 $\mu\text{g em}^{-3}$ in N1 are similar to those in W1, yet $\sigma_{\text{sp,dry}}$ of 70 Mm^{-1} are more akin to W2. E0 features a similar ratio between rBC and $\sigma_{\text{sp,dry}}$ as N1, although the abundance burden is much larger at 1.7 $\mu\text{g em}^{-3}$ and 125 Mm^{-1} .

For all regimes CO mixing ratios are greatest at the near surface: 340 ppb in W1, 310 ppb in E0, 220 ppb in W2 and 150 ppb in N1. CO mixing ratios reduce with altitude to a minimum in the free troposphere: 125 ppb at an unknown altitude in W1, 95 ppb at 4-5 km in E0, 150 ppb at 4-5 km in W2, 100 ppb at 3-4 km in N1, ~~and~~. The reduction of CO mixing ratios in the boundary layer from W1 to W2 corresponds to that of ~~the~~-aerosol abundance burden. CO₂ mixing ratios are also greatest at the near surface: 402 ppm in E0, 393 ppm in N1, 394 ppm in W1 and 397 ppm in W2; representing a particularly prominent where the surface enhancement is particularly prominent, 397 ppm. CO₂ mixing ratios also reduce with altitude with minima of 391 ppm in E0 and N1 or 392 ppm in W1 and W2 at the same altitudes as those for CO.

Profiles of pollutant ratios further illustrate the contrasting aerosol and gas phase vertical distributions (Fig. 9.A-B). Compared with CO and CO₂ ~~the~~-aerosol abundance burden is enhanced at higher altitudes in the boundary layer, even accounting for the removal of plumes (Fig. S3). The peak enhancement is at ~2 km in W2, ~1.5 km in N1, ~4 km in E0 and, in W1, ~2.5 km for $\sigma_{\text{sp,dry}}$ and ~1.5 km for rBC. These distributions are also seen when examining the pollutant ratios within plumes - as altitude increases so does the plume $\Delta\text{aerosol}:\Delta\text{CO}$ ratio (Fig. 9.C-D). We use the term $\Delta\text{aerosol}:\Delta\text{CO}$ to describe both $\Delta\text{rBC}:\Delta\text{CO}$ and $\Delta\sigma_{\text{sp,dry}}:\Delta\text{CO}$ since these aerosol properties co-vary. As these enhancement ratios are typically within the boundary layer, share a common source and in most cases are likely relatively fresh, it is unlikely they are bias by sudden changes in the composition of background air driving the observed gradient, as warned against in (Yokelson et al., (2013).

There is limited evidence for a diurnal variation in aerosol abundance burden and vertical profile shape. CO, however, is enhanced near the surface in the morning (Fig. 8). In W2 and N1, CO₂ is also enhanced at the near surface in the morning but, given the poor correlation with other pollutants (Sect S5), likely arises from biogenic activity. This may also help explain the

greater CO₂ mixing ratios in W2 vs. W1, as the increased cloud cover reduces photosynthesis and hence CO₂ uptake (Graham et al., 2003).

5 Within the boundary layer there is a significant contrast in the coating thickness of rBC between E0 (55 nm) and W1, W2 and N1 (80-90 nm; Fig. 10, Fig. S4). Coating thickness increases with altitude in the boundary layers of W2, N1 and E0. This may be associated with partitioning of organic material into the particle phase as temperatures decrease, and/or the greater prevalence of fresh emissions near surface. In W1, W2 and most markedly N1, the rBC coating thickness declines above the boundary layer. Comparatively, rBC coating thickness increases above the boundary layer in E0.

4 Discussion

10 4.1 Drivers of the pollutant vertical distribution

Fire class determines the relative mix of pollutants emitted. In E0 there are relatively greater loadings of rBC owing to emissions from more efficient flaming combustion in Cerrado fires as observed by Hodgson et al. (2018). In W1 and W2 there are relatively greater magnitudes of σ_{sp_dry} and CO as the higher biomass density and moisture content results in more inefficient smouldering combustion. Significantly, the shift in meteorology between these two phases does not substantially impact the relative abundances of rBC, σ_{sp_dry} and CO to each other.

15 Meteorological conditions ~~determine~~ influence the absolute pollutant concentrations via their impact on removal rates/processes, advection and fire incidence. The driest conditions and greatest fire incidence meant the greatest pollutant concentrations were observed in E0. The reduction in aerosol loadings from W1 to W2 is attributable to a corresponding decline in fire count and increase in wet removal associated with the more widespread and heavy precipitation. Aside from
20 wet removal, the ubiquity of the pollutant residual layer indicates pollutants are not rapidly removed. As such, advection becomes a key modulator of pollutant concentrations local to, and distant from, the source region. The latter is the case in N1 - back trajectory analysis (Fig. S65) shows that pollutants are advected from the fire hotspot around Maranhão and northern Pará states (Fig. 5). The pollutant mix - relatively greater concentrations of rBC and - is consistent with the Cerrado burns at these sources. Whilst CO₂ mixing ratios are also greater in the east (relative to CO), as would be expected from more flaming
25 combustion, the role of fire ~~emissions~~ combustion processes cannot be evaluated given the confounding influence from biogenic sources/sinks in both regions.

The shapes of pollutant vertical distributions are primarily controlled by meteorological conditions, in particular vertical convective motions and horizontal wind shear (Fig. 7). The former acts to mix pollutants released near the surface toward the mixing layer top, the altitude of which can be modulated by the latter, soil moisture and solar insolation. The difference in
30 profile shape from west to east to north is primarily driven by contrasting mixed layer depths. Pollutant loadings remained relatively high above the mixing layer in residual layers, indicating wet removal is not significant at these altitudes. Large unmixed plumes perturbed the mixed and residual layers this state, although they contribute only 15% (E0), 11% (W1), 8%

(W2), and 1% (N1) to the scattering only column AOD (calculated from the nephelometer, Sect. S2). Such plumes were seldom seen above 4 km, in contrast to previous observations in approximately the same sample region (Supplementary material in Andreae et al., 2004), indicating the mass flux from large pyrocumulus detrainment into the upper troposphere (within the aircraft range) was not significant. The observed increases in CO concentrations above ~4km indicate vertical transport of mixing layer pollution into the free troposphere. The presence of co-incident increases in rBC or $\sigma_{sp, dry}$ in less than 10% of these plumes indicates moist deep convection transports CO and presumably other gaseous and non-soluble components to altitudes greater than 4 km but efficiently removes aerosol from the air by wet scavenging. This is consistent with the decrease in rBC coating thickness at these altitudes in W1, W2 and N1 and also is similar to previous observations in Boreal Canada that showed preferential wet deposition of the largest and most coated particles (Taylor et al., 2014). As deep moist convection is not common in eastern regions (e.g. using TRMM rainfall as a proxy, Fig. 4a) the source of elevated and enhanced CO is unlikely to arise from the mixing layer in the east. It is possible the source is from CO aloft in the west which is recirculated in the persistent anti-cyclonic flow at 500 hPa (Fig. S2.f) as has previously been observed from satellite (MOPITT) CO observations by Deeter et al., (2018). A long aging time is consistent with the larger rBC coatings observed aloft in E0.

A transect flight from east to west (Fig. 11) captures and summarises the meteorological drivers of the east-to-west regional contrast in pollutant vertical distribution. A declining mixing layer depth from east to west is evident from the reduction in altitude of the sharp gradient (i.e. the entrainment zone) of θ_e from ~3 km in the east to ~1.5 km in the west. Above the mixed layer top relative humidity increases, especially so after the lifted above the lifting condensation level where the high humidity distinguishes the cloud convective layer. This layer is deeper in the west, associated with deeper moist convection. A wind speed maximum is present at 5-6 km, coincident with the entrainment zone. Together, this structure can explain the lidar derived extinction coefficient distribution (first published by Marengo et al., 2016). Aerosol is capped below the first wind speed jet, well mixed within the mixed layer and features a maximum at ~1.5-2 km. Visible plumes at ~59 °W and ~52.5 °W lie at injection heights typical of those observed in the in situ profile data. The similarity of the in situ $\sigma_{sp, amb}$ and lidar extinction coefficient profile shapes at the regional (Fig. 11) and local (Fig. S6S7) scales engenders confidence in the representativity of both datasets. Disparity in the absolute magnitudes is primarily explainable by differences in the sampling coverage (Fig. S7S8). Substantial dry deposition could remove near surface aerosol and drive the observed Δ aerosol:ACO. However, fluxes reported in the literature (Ahlm et al., 2010) are small compared to the number concentrations observed (Darbyshire et al., 2018) and thus we discount the role of dry deposition. CO near surface enhancements in the morning indicate smouldering emissions overnight may be particularly significant as pollutants accumulate in the nocturnal boundary layer.

~~— Plume injection altitude is potentially significant in driving the contrasting vertical distributions between different pollutants in all regimes. We observe the relationship between Δ aerosol:ACO and altitude is driven by plumes at higher altitude being enriched in aerosol and at low altitudes enriched in CO. We speculate the plumes at higher altitude arise from fires with more efficient combustion and at lower altitudes from less efficient combustion. This is consistent with the relationship described between combustion efficiency and Δ aerosol:ACO from previous observations in the region (Fig. S8; Ferek et al., 1998; Yokelson et al., 2007) and SAMBBA (Fig. S9, Hodgson et al. 2018). Previous studies of biomass burning plumes have~~

described the relationship between combustion completeness phase plume injection height (Akagi et al., 2011; Freitas et al., 2006). As such, we postulate the enhanced aerosol at altitude is due to lofted emissions from more flaming combustion, and the reduced aerosol near surface results from a dominance of smouldering emissions. CO near surface enhancements in the morning indicate smouldering emissions overnight may be particularly significant as pollutants accumulate in the nocturnal boundary layer. Substantial dry deposition could remove near surface aerosol and drive the observed Δ aerosol: Δ CO. However, fluxes reported in the literature (Ahlm et al., 2010) are small compared to the number concentrations observed (Darbyshire et al., 2018) and thus we discount the role of dry deposition.

Consistent values of the scattering angstrom exponent at 700/550 nm within the boundary layer (Fig. S5) indicate a similar aerosol type throughout. These values were all above 1.5, typical of submicron biomass burning aerosol and indicating no significant regional role for super-micron dust or primary biological aerosols which have values closer to zero (Clarke et al., 2007; Russell et al., 2010). This is consistent with size distributions reported and from the aircraft (Darbyshire et al., in preparation). This also indicates that model scaling factors to match remotely sensed AOD are not significantly biased by non-biomass burning aerosol. The observed increase in humidity with altitude in the boundary layer (Fig. 7C) will likely have a significant impact on AOD and therefore the required scaling factor as it remains an uncertain model process (Johnson et al., 2016; Reddington et al., 2018). We note that findings from SAMBBA suggest that omission of burned area from small undetected fires is the most significant source of inventory under-representation of aerosol emissions (Hodgson et al., 2018; Reddington et al., 2016b), as in other burning regions (Nowell et al., 2018).

The average vertical distribution of pollutants in this study are similar to those from previous in situ and remote sensing observations in the region, despite temporal, geographic and instrumental sampling differences. This indicates driving processes are common throughout. The aerosol vertical profile is approximately vertical from the surface to a height of: 2.25 km (Huang et al., 2015; Table 2 therein), ~1.5 km (Bourgeois et al., 2015; Fig. 6 therein), ~2 km (Baars et al., 2012; Fig. 12 therein), ~1.5 km (Andreae et al., 2012; Fig. 6 therein), ~2 km (Marenco et al., 2016; Fig. 9 therein) or 1.5 - 3.5 km (this work, Fig. 8). Loadings reduce to near baseline values above ~4 km in all. Only in Andreae et al. (2012) have similar regionally averaged profiles of CO been presented. These exhibit a similar profile shape as here - greatest near the surface and reducing thereafter to a minima at ~4 km. Andreae et al. (2012) This study therefore also demonstrated the same relationship of Δ aerosol: Δ CO with altitude as observed here. Whilst studies displaying only a few profiles (e.g. Andreae et al., 1988, 2004; Chand et al., 2006; Pereira et al., 1996; Yokelson et al., 2007) may not mirror these average distributions, comparable profiles are present in our library of individual profiles (Sect S5;) indicating that our more extensive dataset is consistent with previous reports and captures variability between different studies.

To try understand the relationship between Δ aerosol: Δ CO and altitude it is necessary to examine multiple potential drivers. Substantial dry deposition could remove near surface aerosol and reduce the observed Δ aerosol: Δ CO. However, dry deposition fluxes reported in the literature (Ahlm et al., 2010) are small compared to the number concentrations observed (Darbyshire et al., in prep) so this is an unlikely cause. Likewise we discount a significant anthropogenic non-biomass burning source of CO near the surface, as emissions hotspots were over 1000 km from the flight region and are approximately an order of magnitude

lower than from fires (Fig. S9). Increasing aerosol concentrations with altitude are typically attributed to partitioning of organic and inorganic species from lower volatility gas phase species into the particulate phase as temperatures decline and relative humidity increases (Heald et al., 2011; Morgan et al., 2010). A significant fraction of boundary layer organic aerosol in the regional haze will be secondary in nature following oxygenation and condensation of semi-volatile vapours during smoke plume evolution. During SAMBBA no net addition of organic mass was observed in the near or far field (Morgan et al., 2019), unlike previous studies (e.g. Akagi et al., (2012)). However, the contribution of further biomass burning secondary organic aerosol owing to increasing altitude is difficult to determine with the data available. There is an increase with altitude of the contribution from secondary inorganic aerosol with altitude in E0, but not W1, W2 or N1 (Darbyshire et al., in prep). This may account for the increase in rBC coating thickness with altitude (Fig. 10), but the decrease with altitude in W1, W2 and N1 indicates no significant addition of secondary organic or inorganic aerosol. Furthermore, as the approximate three-fold increase of involatile $\Delta rBC:\Delta CO$ with altitude is of the same magnitude to that of $\Delta sp:\Delta CO$, these secondary aerosol processes cannot explain our observations.

Plume $\Delta aerosol:\Delta CO$ enhancement ratios increase with altitude within the boundary layer, indicating that these plumes may be the source of the observed $\Delta aerosol:\Delta CO$ profiles in the regional haze. Past measurements (Fig. S10; (Ferek et al., 1998; Yokelson et al., 2007) and those from SAMBBA (Fig. S11, (Hodgson et al., 2018) indicate $\Delta aerosol:\Delta CO$ increases with MCE. We speculate the distribution may be driven by the relationship between MCE and fire intensity – and thus by extension, when normalising for meteorology, plume height (Lavoué et al., 2000). The peak heat flux (i.e. intensity) of an open landscape fire occurs when there is both a flame front, where flaming combustion occurs (high MCE), and zone of smouldering combustion (moderate MCE) in its wake. Once the flame front is extinct the fuel bed remains smouldering (moderate MCE) and although reduced, there may still be sufficient heat flux to generate a convective column to loft these emissions, albeit to intermediate altitudes. Once the heat flux is insufficient to generate any convective plumes and emissions from residual smouldering (low MCE) are released at the surface. Together one would thus expect a continuum from high altitude release of high+moderate MCE emissions to surface release of low MCE emissions, which could explain our $\Delta aerosol:\Delta CO$ observations. The evolution of plume height as a function of heat flux has previously been observed for savannah fires in South Africa (Stocks et al., 1996). Alternatively, there may be multiple convective cores for a given burn as seen elsewhere (Achtmeier et al., 2011). Where these are away from the primary flame front the MCE will be lower along with heat flux and therefore injection height. Another complicating factor may be from very intense fires which experience oxygen deficiency in the flaming zone and combustion efficiency drops (Ward and Hardy, 1991) – and therefore aerosol:CO reduces. We recommend further fire-scale plume dynamics observations in tropical regions to test these speculations and acknowledge that many case studies would be required to account for the other impacts on ‘real world’ fires plume rise (including stability, vertical profile of wind speed/direction, latent heat release, flame front annihilation, topography).

In addition to being related at the individual fire level, regionally there is a connection between MCE, average fire intensity and injection height. Fires in eastern Cerrado regions have a higher MCE (Hodgson et al., 2018), are more intense as illustrated

by the FRP distributions in Fig. 5b and (Gonzalez-Alonso et al., (2018), and have higher plume injection heights in spite of more stable conditions (Fig. S12, (Gonzalez-Alonso et al., 2018; Marengo et al., 2016).

4.2 Implications

We have observed a marked contrast in pollutant composition and vertical distribution from west to east across tropical South America which has not previously been identified. Given predicted ‘savannisation’ of the southern and eastern edges of the Amazon rainforest owing to climate change, deforestation and fire-climate feedbacks (Nobre et al., 2016) we recommend model analyses are carried out to assess the magnitude of these effects which may impact convective motions, cloud formation and regional dynamics.

The linkages between pollutants and atmospheric structure presented here reinforce the first order requirement for representation of realistic thermodynamics within models. This is currently a challenge for regional and global models, especially sub-grid convective mixing which drives the aerosol profile shape (Archer-Nicholls et al., 2016; Hong and Dudhia, 2012). For instance ECMWF ERA-Interim equivalent potential temperature profiles compare poorly to our measurements (Fig. 11) and those of Beck et al. (2013). These model fields are often used as boundary conditions for regional models and as such may introduce unrealistic thermodynamic structures and thus pollutant vertical distributions. Whilst the broad shape of horizontal wind shear from model output is captured on the individual profile basis, the fine structure present in observations is missing. Comparisons with ECMWF ERA-Interim and WRF output indicate this is the case for both global models and finer scale regional models. This may be problematic in accurately representing sub-grid processes, such as plume rise and convective mixing.

We speculate the Δ aerosol: Δ CO profile may be driven by a coupling between combustion efficiency and plume dynamics. If confirmed by further enquiry then future modelling studies will have to consider how best to represent the phenomenon. Replicating the gradient Δ aerosol: Δ CO may be particularly important for Following ignition a fire undergoes a period of more complete combustion, the flaming phase, a period of less complete combustion, the smouldering phase, and then a period of even less complete combustion, the residual smouldering phase. Based on our results we hypothesise these less prominent phases are potentially important in driving the observed Δ aerosol: Δ CO profile. Aerosol rich emissions from the initial flaming phase will be lofted, whilst CO rich emissions from residual smouldering will be released near surface. Convective mixing will act to rebalance this contrast, although it remains owing to the ubiquity of residual layers and plume injections aloft. Past satellite observations have suggested residual smouldering is a large source of CO and not fully captured by emissions inventories (Deeter et al., 2016; Pechony et al., 2013), which may be based on inappropriate emissions factors or arise from small fires outside the detection limit of active fire detection schemes (Giglio et al., 2016; Schroeder et al., 2008). Even if single fire integrated emissions factors represent the total pollution released, our results suggest their use may not always be appropriate in model simulations which draw results based on a realistic vertical distribution. For example aerosol-cloud and aerosol-radiation interactions, or surface air quality simulations. Emissions factors for residual smouldering combustion in the Amazon have been collected (Bertschi et al., 2003; Christian et al., 2007), and could be used to test predictions from novel

(and non-trivial) model set-ups against surface and satellite observations. Past satellite observations have suggested residual smouldering is a large source of CO and not fully captured by emissions inventories (Deeter et al., 2016; Pechony et al., 2013).

If emissions from higher MCE flaming combustion are ~~to be~~ released at altitude, further exploration of the processes and variability in TSA is required. Archer-Nicholls et al. (2015) demonstrated how the parameterised Freitas et al. (2007) plume rise model calculated injection heights which were too high in TSA. Climatological distributions of plume injection heights, which can prescribe model injection heights, show substantial diversity in TSA. The distributions derived by Marengo et al. (2016), consistent with plume heights in this study (Fig. S10), show injection heights at greater altitudes than those from CALIOP (Sofiev et al., 2013), and in turn MISR (Gonzalez-Alonso et al., 2018) and MISR (Fig. S11) satellite observations.

Although the magnitude and bearing may differ, the fundamental drivers of the pollutant vertical distribution identified here will remain so in drought years which may be increasing in frequency (Jiménez-Muñoz et al., 2016). Dry convection may be more vigorous and the atmosphere more stable, deep convection less vigorous and aerosol scavenging reduced, fires more intense and fire hotspots located in different regions, but so long as model simulations well represent the fundamental drivers identified in this work then they ought to be able to replicate the resultant vertical distribution. This is a promising avenue for future research to predict the impacts in future years, following on from the study of (Thornhill et al., (2018).

15 **6 Conclusions**

The vertical distribution of biomass burning pollutants regulates their subsequent horizontal distribution and thus the magnitude of associated impacts on weather, climate and air quality. In tropical biomass burning regions, pollutant vertical distributions and their driving processes are poorly characterised. Here, a novel integrated analysis of individual profiles from research flights in tropical South America will aid future experiments in~~has helped reduce- reducing this some of these~~ uncertainties.

The thermodynamic structure of the lower troposphere was found to be critical in determining the pollutant vertical distribution on the local and regional scale. Pollutants were typically confined to the atmospheric boundary layer in either an active or residual convective mixed layer. This was deeper in the east (~3 km) than west (~1.5 km) reflecting a regional contrast in soil moisture and surface insolation and as such significant differences in sensible and latent heat. Above the boundary layer, enhanced concentrations of CO were likely transported via deep moist convection which removed aerosols via wet scavenging. Horizontal wind shear was important in advecting pollutants, inhibiting mixed layer growth and the vertical ascent of smoke plumes from fires.

In order to accurately simulate the vertical and regional distribution of biomass burning pollution our results highlight not only the importance of capturing boundary layer dynamics and convection adequately, but also the release of pollutant plumes at altitude.

During dry season conditions we observed a significant contrast in the pollutant haze composition between the western and eastern regions, which corresponded to different fire regimes. Whilst aerosol scattering coefficients of 120 Mm^{-1} and CO

mixing ratios of ~310 ppb were similar, black carbon mass loadings were much greater in the east than west - 1.5 vs. 0.85 $\mu\text{g em}^{-3}$. Cerrado burns in the east are more flaming, whilst those in the west, of primary/secondary forest and pasture land are more smouldering (Akagi et al., 2011; Andreae and Merlet, 2001; Hodgson et al., 2018). The is discrepancy regional contrast in biomass burning emissions resulting from these different fire types clearly merits future investigation in modelling studies

5 to assess potential impacts on aerosol optical properties and their radiative effects.

Following the transition to wet season conditions in the west, fire activity declined and the observed concentration of black carbon mass, aerosol scattering coefficients and CO mixing ratios decreased. However, the ratios between these pollutants remained similar to ratios observed in the drier phase indicating little change to the composition of fire emissions despite the shift in meteorological conditions.

10 In the remote rainforest region of northern Amazonas state, significant concentrations of well mixed and thickly coated black carbon particles were observed in the lowermost 2 km and with comparatively low aerosol scattering coefficients. Trajectory analysis indicated origins from north-east Brazil where Cerrado fires were prevalent and, based on observations of Cerrado fires elsewhere, likely emit aerosol with a relatively high BC mass fractions. Given the high fire count and fire radiative power in this source fire region and the prevailing low level easterlies, the export of absorbing aerosol into the remote
15 central regions of Amazonia may commonly occur during the dry season.

~~—In all regimes we observed an increasing contribution of pollutants from more flaming combustion with altitude—there is relatively more aerosol aloft and more CO near the surface. Typically there is no explicit representation in atmospheric models of emissions from only the residual smouldering stage, which would remain near the surface, nor those only from the flaming stage, which would also require transportation to an appropriate injection altitude. This may impede accurate predictions of aerosol radiation and aerosol cloud interactions, in addition to air quality forecasts. Further enquiry is recommended to test the magnitude of these findings.~~

20 In all regimes we observed an increase of aerosol abundance relative to CO with altitude both in the background haze and plume enhancement ratios. This is unlikely associated with thermodynamic partitioning, as the phenomenon is observed for involatile rBC, nor surface deposition or non-biomass burning sources. Aerosol abundance relative to CO also varies with combustion efficiency, and we speculate that this may drive the observed gradient as combustion efficiency and plume injection height can be closely coupled. If the observed gradient is not fully captured by model simulations this may impede accurate air quality forecasts and predictions of aerosol-radiation or aerosol-cloud interactions. . Further enquiry is recommended to fully explain these observations and explore the ramifications for regional climate and air quality. .

25 The results here ~~may be~~ likely applicable to other tropical biomass burning regions where observations of processes affecting pollutant vertical distributions are limited. Given the important impacts of biomass burning on meteorology, climate, air quality and ecosystem services, further vertically resolved observations of aerosol and trace gas pollutants are recommended.

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Data Availability

All raw time series data used to derive the vertical profiles from the FAAM research aircraft are publicly available from the Centre for Environmental Data Analysis website (<http://www.ceda.ac.uk/>, last access: 31 August 2018). A direct links to the flight data records are given in the reference list (Facility for Airborne Atmospheric Measurements, Natural Environment Research Council, and Met Office, 2014). Raw active fire and Land use data used in the manuscript are available publicly from NASA and ESA respectively (see Acknowledgements). Processed individual and averaged vertical profiles, data masks, plume composition, model output and satellite fields and are currently available on request from E. Darbyshire. Lidar data is available on request from F. Marengo (franco.marengo@metoffice.gov.uk).

Author Contributions

ED analysed the data and wrote the manuscript with the aid of WTM, HC, BJ, JFB and FM. WTM, JDA, DLiu, DLowe, SO'S, FM and KS provided additional data analysis support, including data processing and quality assurance. MJF, JD, KS and JFB operated aerosol instruments during the field campaign. SB operated gas phase instruments during the field campaign. BJ, JMH, KML, PA and HC led the planning of the field campaign and were co-principal investigators on the SAMBBA project.

15 Competing Interests

The authors declare that they have no conflict of interest.

Special Issue Statement

This article is part of the special issue “South American Biomass Burning Analysis (SAMBBA)”. It is not associated with a conference.

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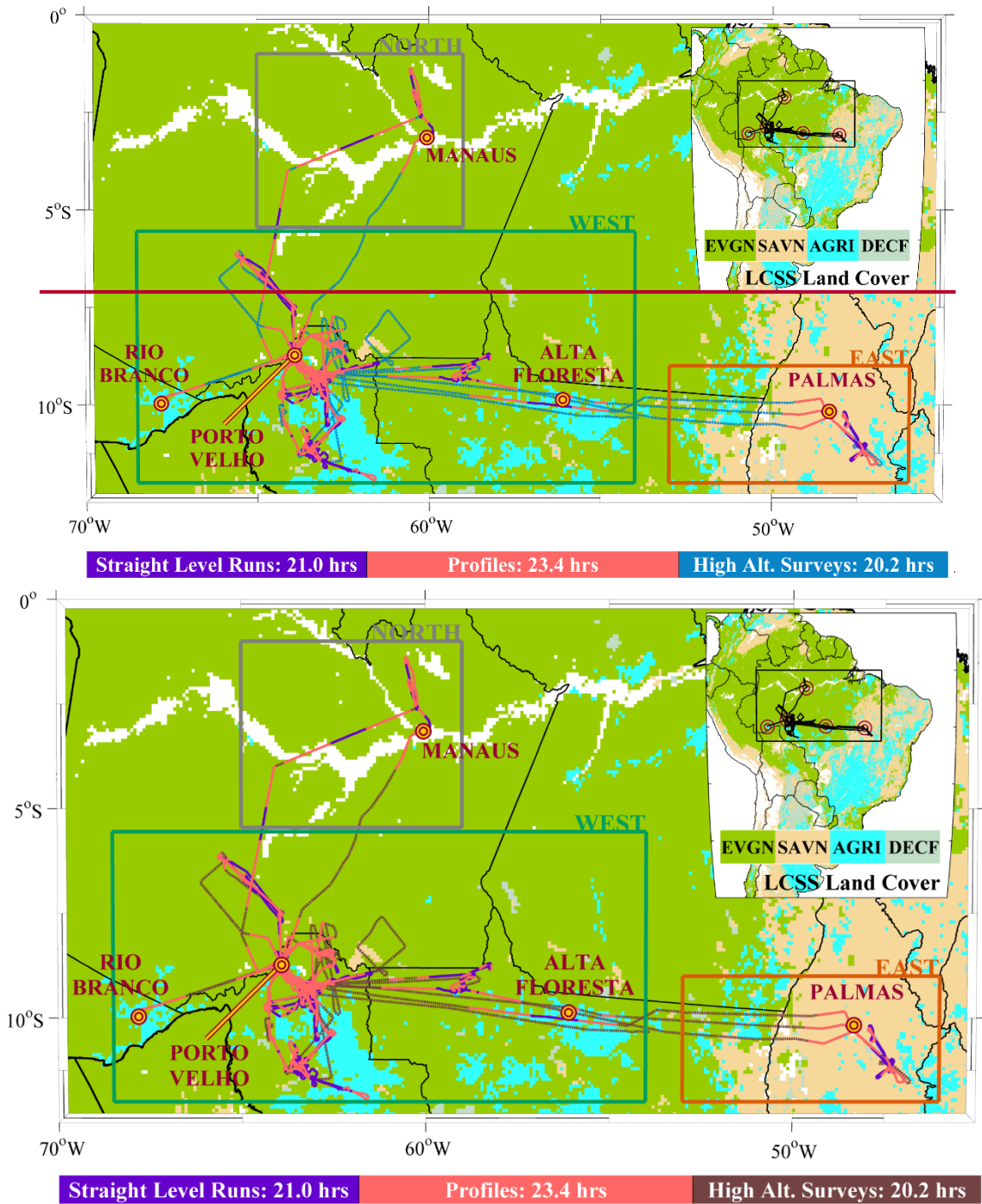


Figure 1. SAMBBA operational domain with flight tracks coloured by basic aircraft operation. 2012 land use based on a simplification of the UN Land Cover Classification System (LCCS) classification as described in the supplementary information: evergreen forest (EVGN), savanna (SAVN), deciduous forest (DECF) and agricultural land (AGRI).

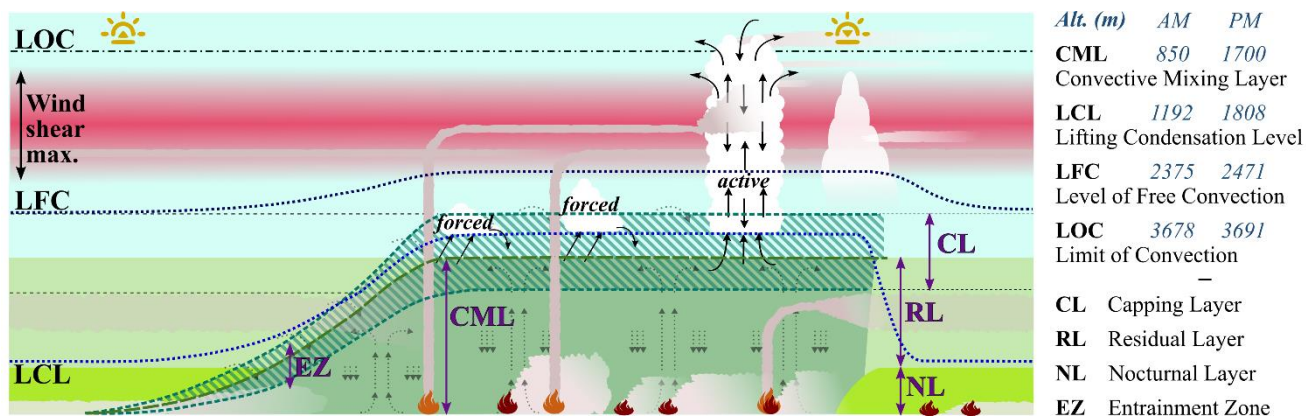


Figure 2. Schematic of the typical diurnal development of the convective boundary layer. This is based on visual and automated analysis of individual profiles and, for the nocturnal period outside our sampling times, previous tropical boundary layer literature. Red flame symbols indicate more smoldering fires and orange symbols more flaming combustion.

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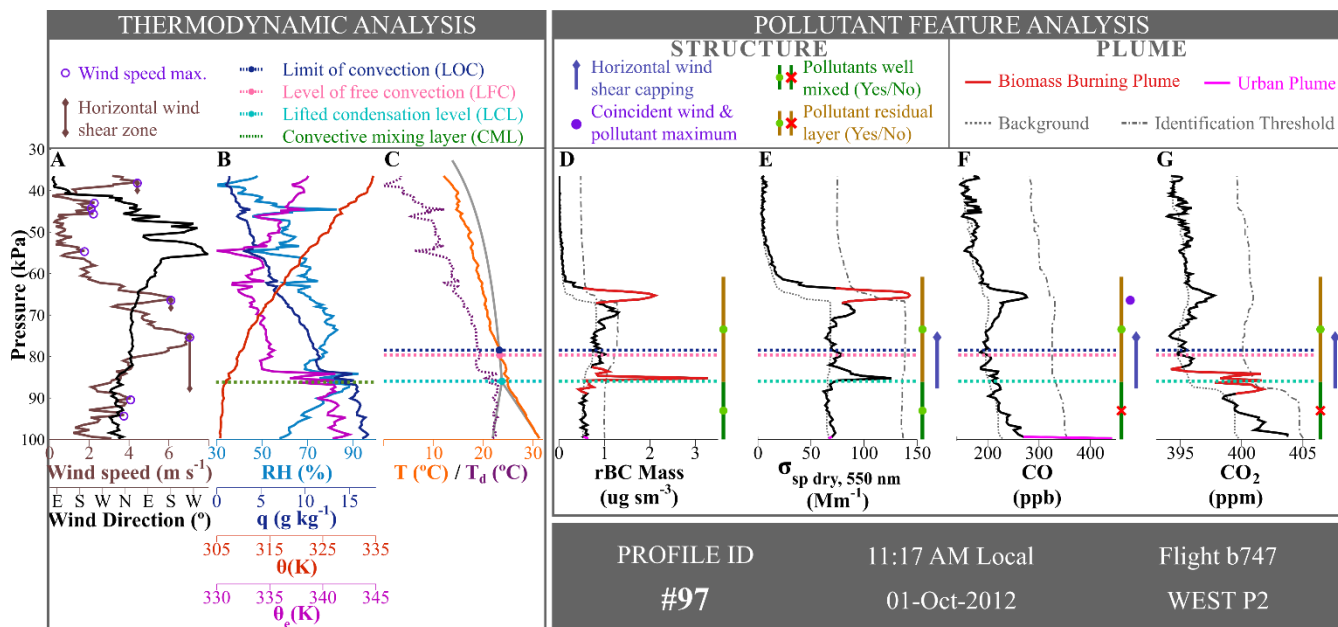


Figure 3. Individual profile #97, from flight b747 (01/10/2012) near Porto Velho in Rondônia state. From left to right are the vertical profiles of (A) wind speed and direction, (B) relative humidity (RH), specific humidity (q), potential temperature (θ) and equivalent potential temperature (θ_e), (C) temperature (T) and dew point temperature (T_d) on a skew-T log-P scale, (D) refractory black carbon (rBC) mass, (E) aerosol scattering coefficient (550 nm), (F) CO and (G) CO₂ mixing ratios. Thermodynamic features are illustrated in panels A-C (see Sect. 3.1). Pollutant feature analysis is illustrated in panels D-G (see Sect 3.2). **Note, the red outline of a biomass burning plume is only present if it was identified for that specific species. For example at 65 kPa rBC and $\sigma_{sp, dry}$ pass the identification threshold (grey dot-dash line) but CO and CO₂ do not.**

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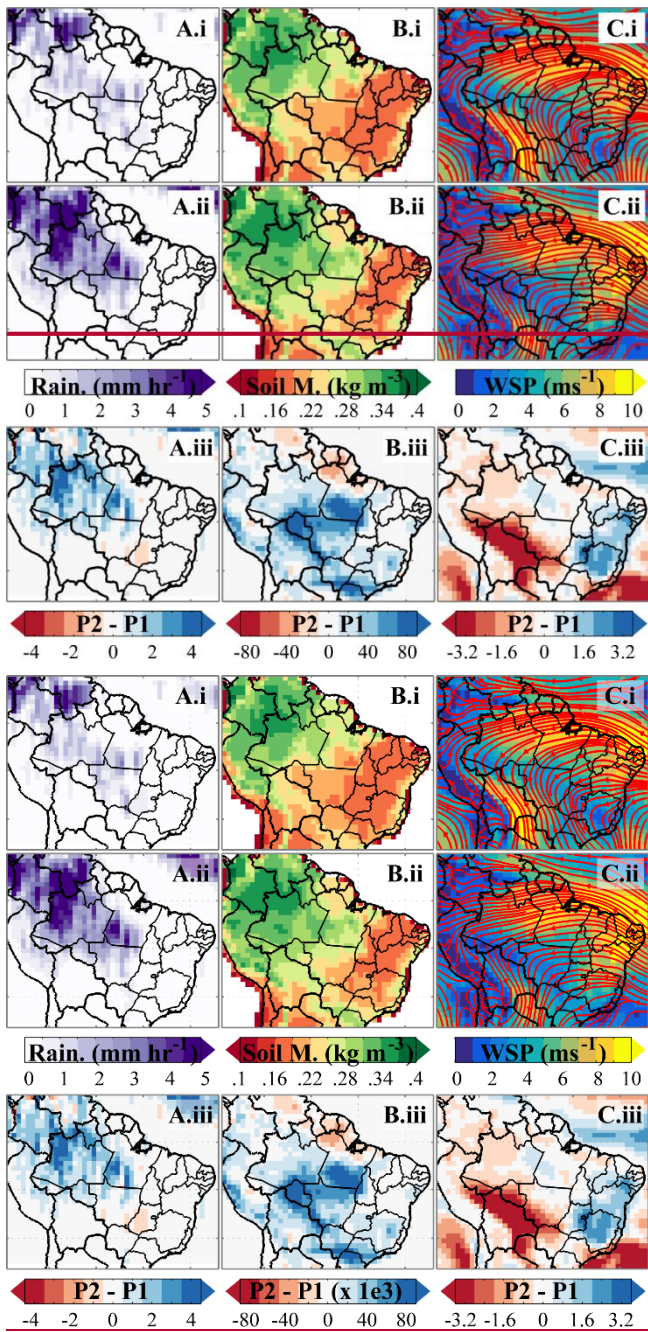


Figure 4. Precipitation (A), soil moisture (B) and 850 hPa wind speed and flowlines (C) during meteorological phases 1 (i, dry season) and 2 (ii, dry-wet transition season) and the difference between the two periods (iii). Precipitation is derived from TRMM measurements. The soil moisture and wind speed products are derived from ECMWF Era-Interim product.

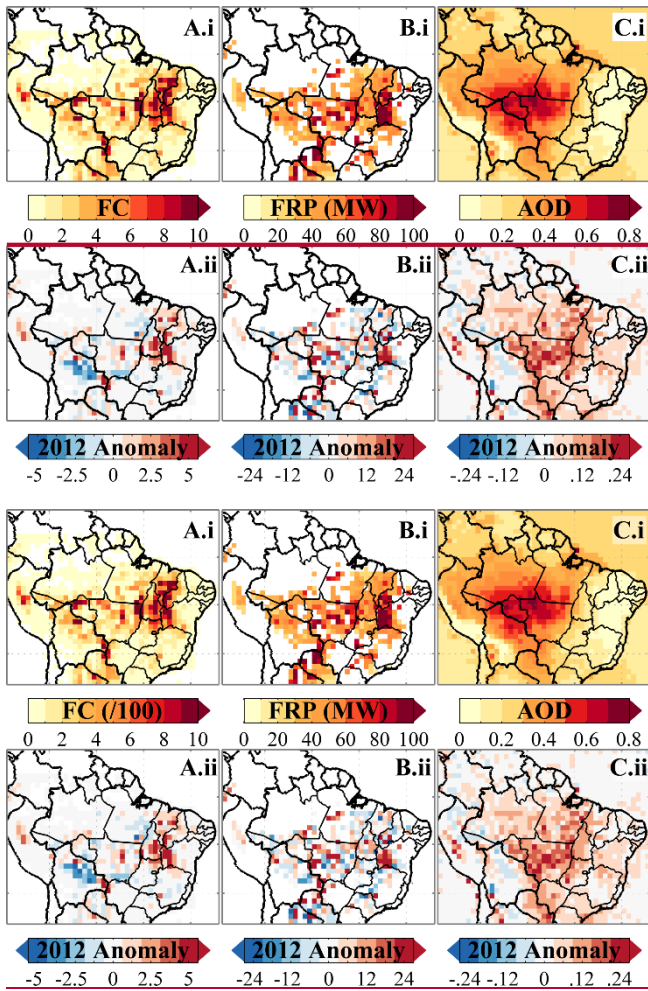


Figure 5. September 2008-2017 median total fire count (FC; A) and fire radiative power (FRP; B) from MODIS active fire detections, calculated over a 1° grid. The FC scale is divided by 100 for figure clarity. September 2008-2017 median aerosol optical depth (AOD; C) from MODIS 1° product. The 2012 anomaly for each of these fields is illustrated in panels (ii).

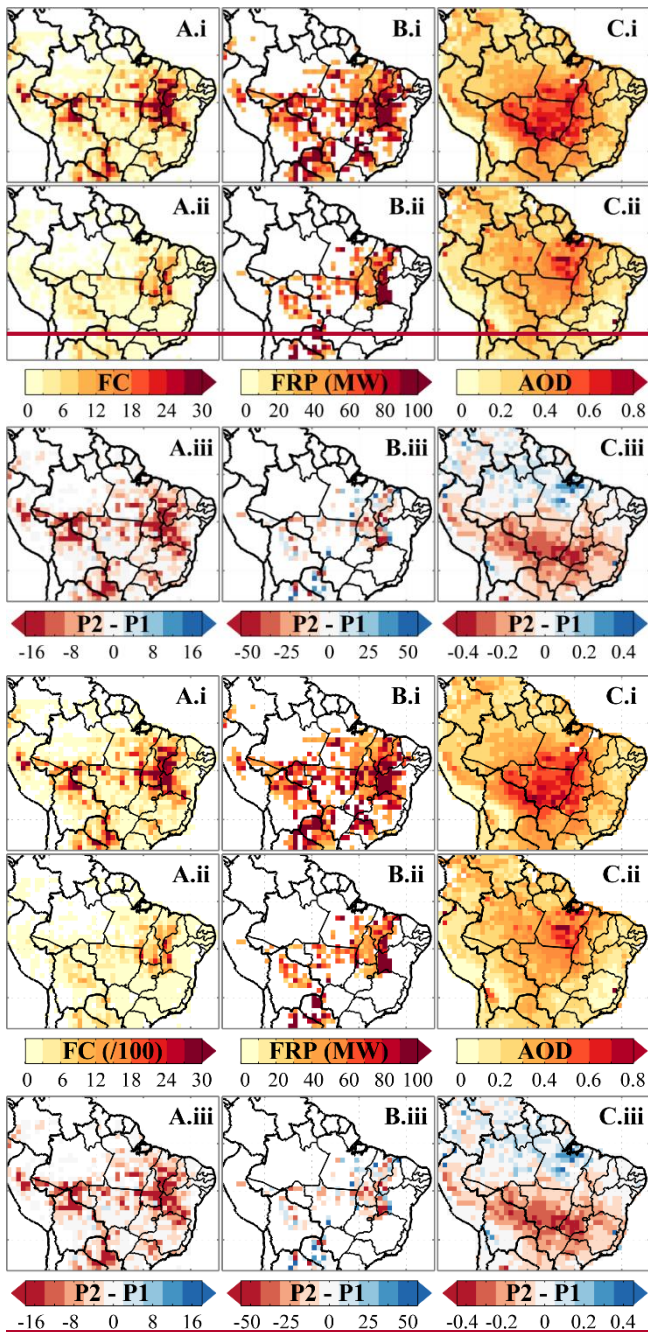


Figure 6. Meteorological phases 1 (i; dry season 14th-22nd Sept 2012) and 2 (ii; dry-wet transition 23rd-5th Oct 2012) median total fire count (FC; A) and fire radiative power (FRP; B) from MODIS active fire detections, calculated over a 1° grid. Meteorological phases 1 (i; dry season) and 2 (ii; dry-wet transition) median aerosol optical depth (AOD; C) from MODIS 1° product. The difference between these periods is illustrated in panels (iii).

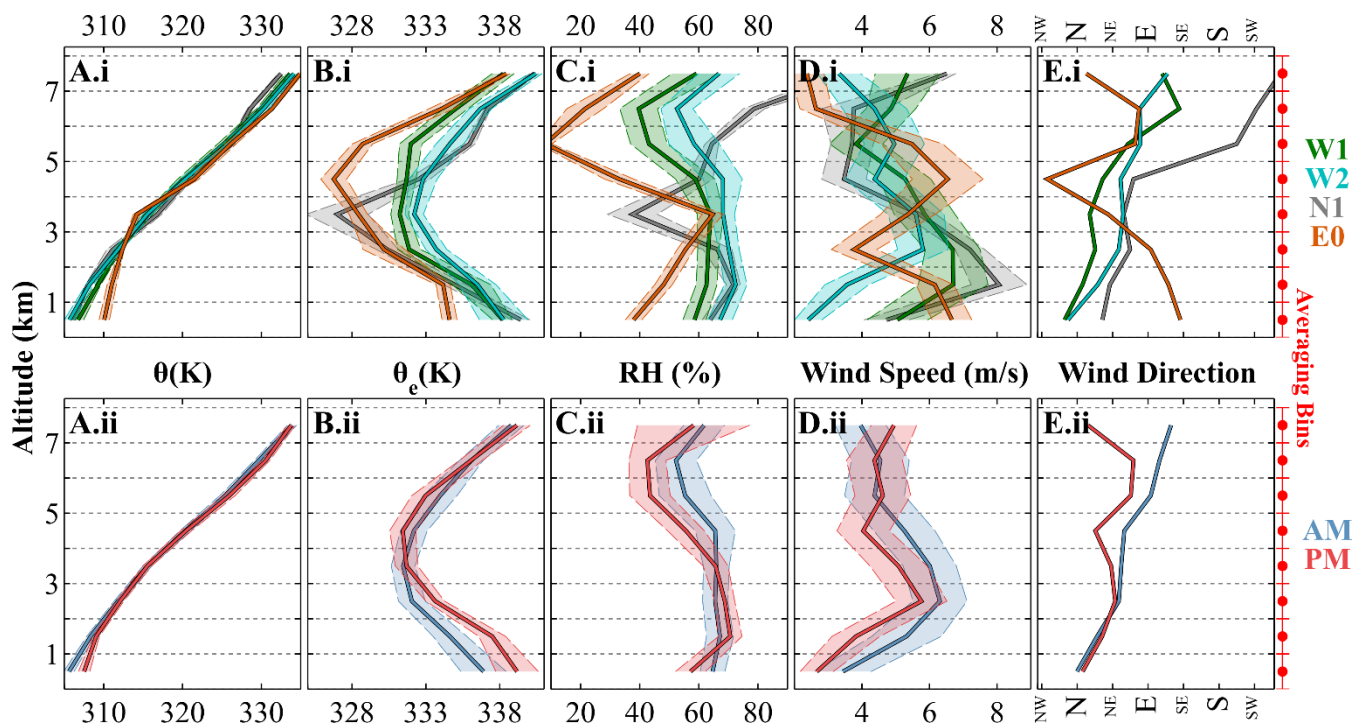


Figure 7. Median pollutant profiles averaged over the different regimes (top panels) and time of day (bottom panels, includes data from all regimes). Lighter shading represents the median absolute deviation.

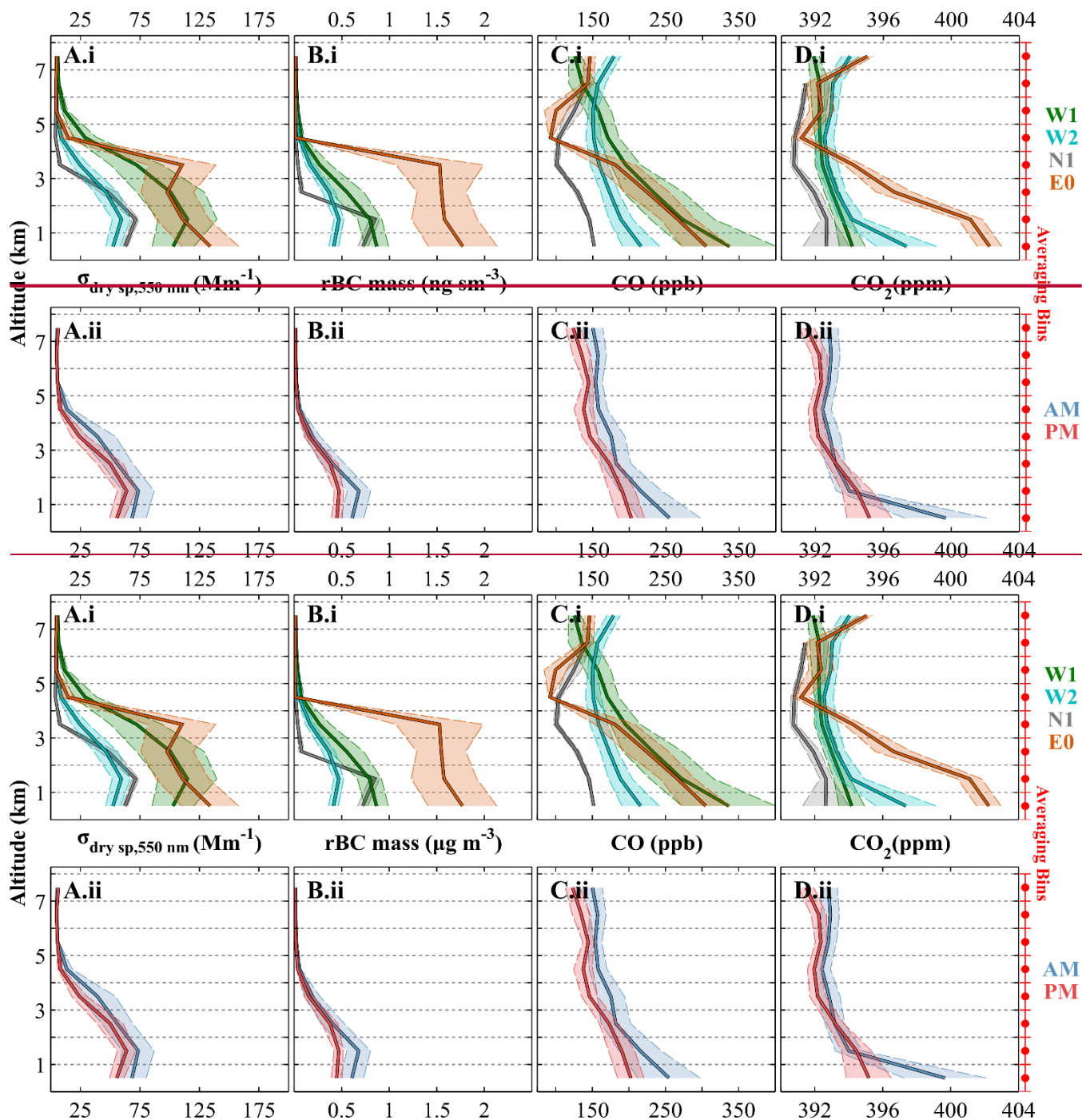


Figure 8. Median pollutant profiles averaged over the different regimes (top panels) and time of day (bottom panels, includes data from all regimes). Lighter shading represents the median absolute deviation. Note, $\sigma_{sp,dry}$ is reported at standard temperature, standard pressure and 30% RH and hence the column AOD cannot be derived from these profiles.

5

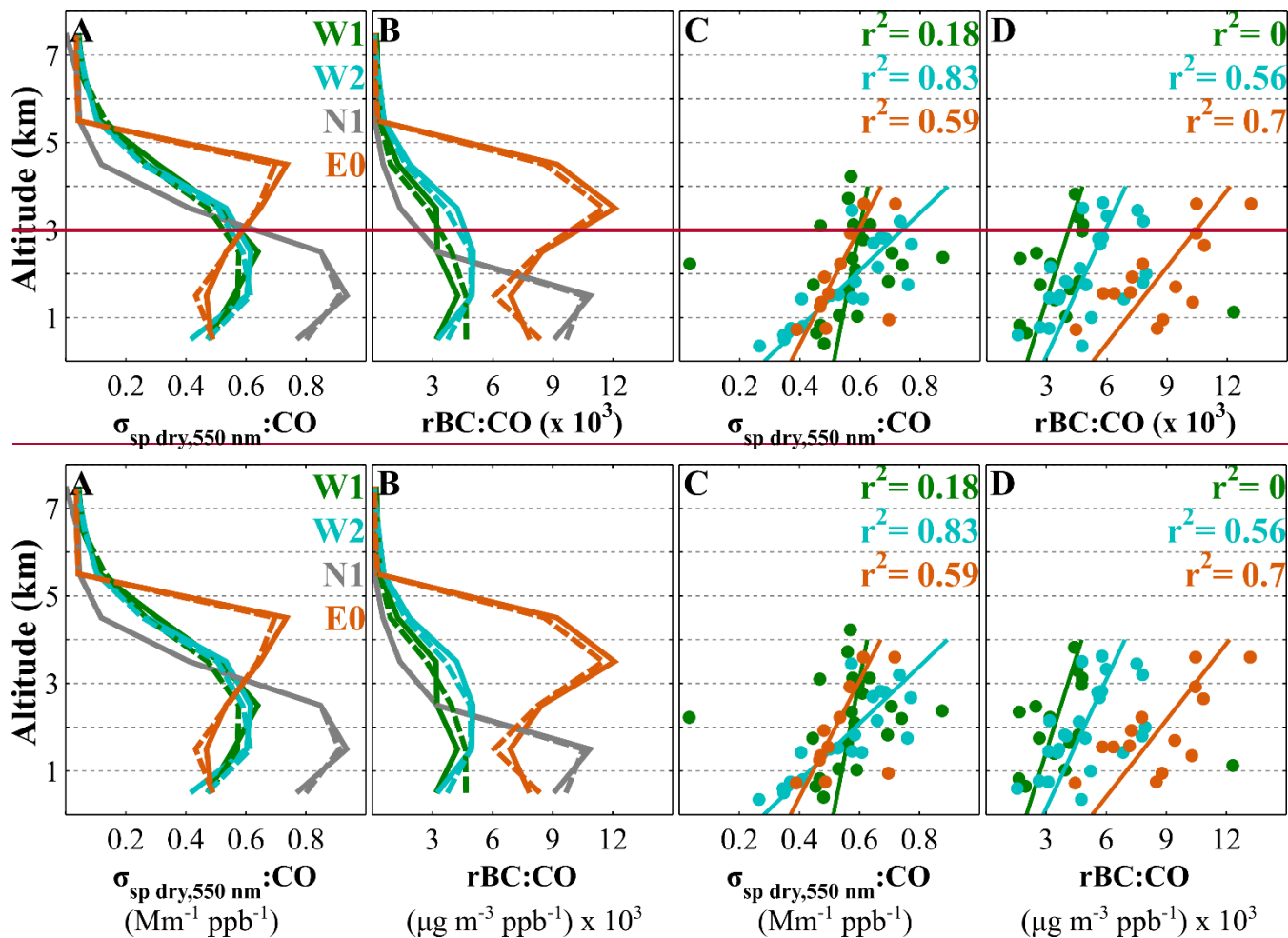


Figure 9. Median enhancement profiles of $\sigma_{sp,dry}:CO$ (A) and $rBC:CO$ (B) enhancement profiles for each regime. Solid (dashed) line represents averages with (without) plumes included. The enhancements were calculated for each individual profile by subtracting the regime 5th percentile. (A,B), and of plumes (C,D). Profile enhancements calculated from data minus the regime 5th percentile. Plume enhancement ratios of $\sigma_{sp,dry}:CO$ (C) and $rBC:CO$ (D) calculated from plume integrated values above the local background (moving 5th percentile, i.e. brown dashed grey baseline Fig. 3) only when the two pollutants are well correlated (Pearson's $r > 0.5$). Ratios reported for pollutants are at ambient temperature and pressure. Solid (dashed) line represents averages with (without) plumes included. Plume enhancements calculated from plume integrated values above the local background (moving 5th percentile, i.e. brown baseline Fig. 3) only when the two pollutants are well correlated (Pearson's $r > 0.5$).

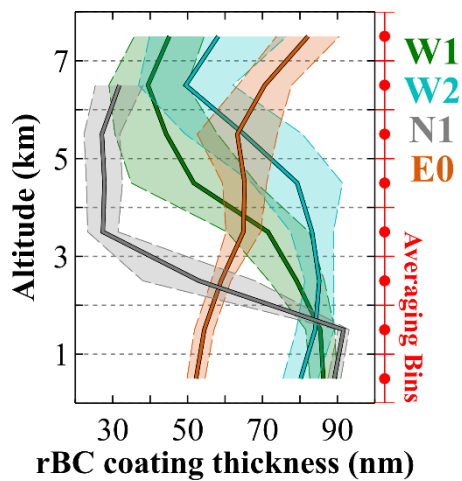
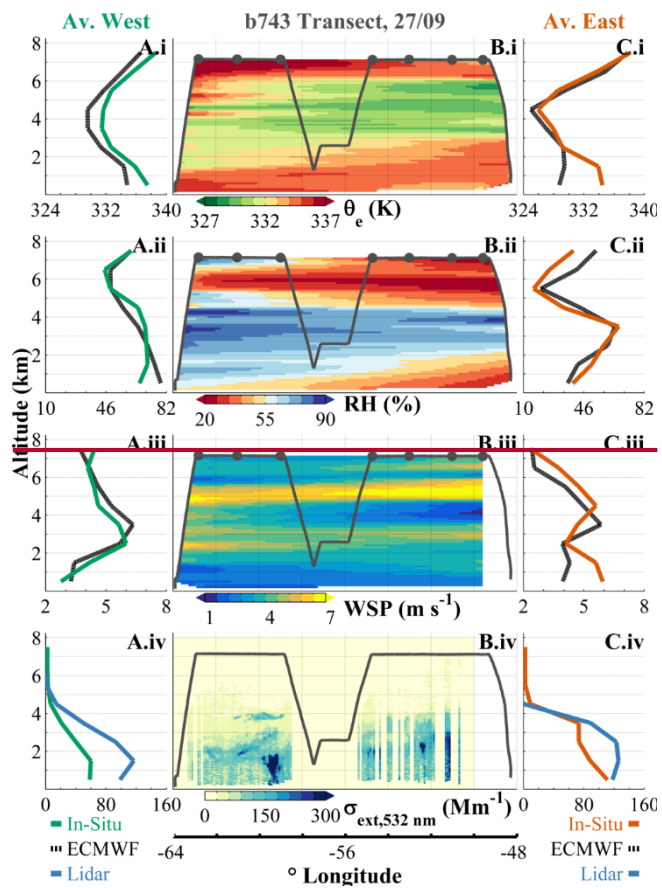


Figure 10. Regime median profiles of black carbon coating thickness. Lighter shading represents the median absolute deviation.



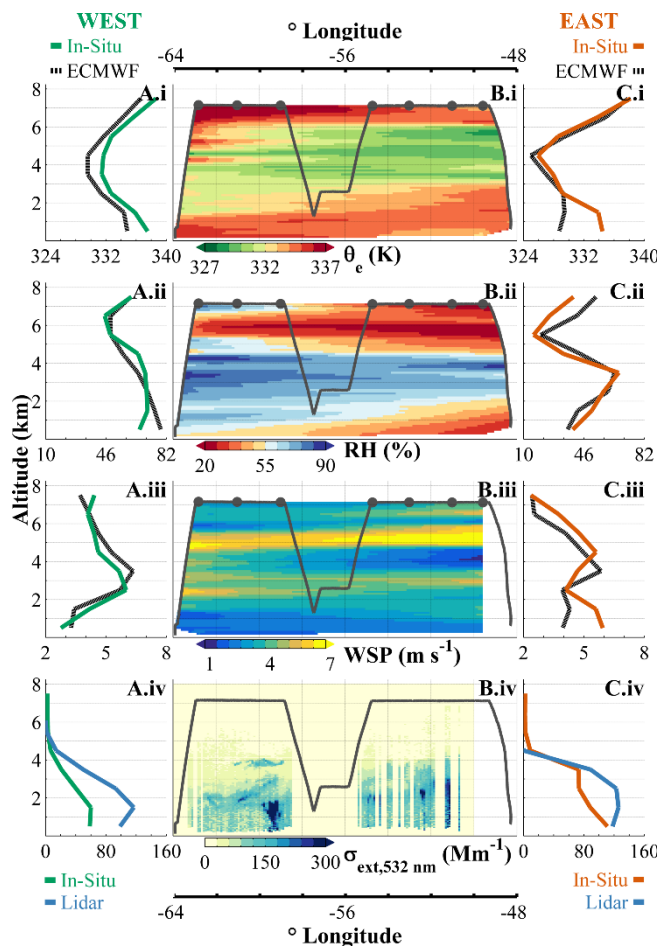


Figure 11. Summary of west to east regional gradient in equivalent potential temperature (i), relative humidity (ii), horizontal wind speed (iii) and aerosol extinction (iv) based on an east to west transect (Panels B) on the 27th September 2012 (flight b743). The thermodynamic curtains (B.i-iii) are derived from linearly interpolated aircraft (grey line) and dropsonde (grey dots) profiles. The curtain of the lidar extinction coefficient at 532 nm (B.iv) is reproduced from Marengo et al., (2016) scaled from 355nm using a scaling factor of 0.57 following Marengo et al., (2014). The regional gradient is emphasised by the side panels which show the median thermodynamic and aerosol parameters extinction profiles averaged (median) across for all western (A) and eastern flights (C). In panels (i) to (ii) the observed thermodynamic parameters are compared to ECMWF reanalysis data extracted along the flight paths. In panel (iv) the in-situ aerosol scattering coefficient at 550 nm is compared to the lidar extinction coefficient averaged over a similar region (Fig. S6; n.b. both measurements are reported at ambient temperature and pressure).

We thank the anonymous reviewer for her/his thorough evaluation and constructive recommendations for improving this manuscript. Her/his comments (in italics) and our responses are listed below.

This manuscript reports findings from the 2012 SAMBBA field campaign with respect to atmospheric distributions of several key pyrogenic pollutants (smoke aerosol, black carbon, and CO). Going further, the manuscript interprets the effects of meteorology on the observed distributions. Much of the detailed information with respect to the data and methods is provided in a Supplementary file. Overall, the paper is scientifically sound and well written, and should be interesting to many readers of ACP. Most of the suggested revisions listed below are relatively minor.

The only significant objection I have to the content is the lack of any consideration of the effects of deep convection on the transport of pollutants in the Amazon Basin. While this mechanism may become more significant towards the dry/wet transition season, a number of studies have reported finding elevated CO concentrations in the Amazonian upper troposphere, likely caused by deep convection. Among these are Andreae et al., 2001 (Geophys. Res. Lett., 28(6), 951–954, <https://doi.org/10.1029/2000GL012391>), Livesey et al., 2013 (Atmos. Chem. Phys., 13, 579–598, <https://doi.org/10.5194/acp-13-579-2013>), and Deeter et al., 2018 (J. Geophys. Res., 123, <https://doi.org/10.1029/2018JD028425>).

In fact, a significant number of the CO profiles presented in the author-provided Supplementary file exhibit features consistent with vertical transport via deep convection. For example, roughly between a third and a half of the 'complete' SAMBBA CO profiles (including CO measurements in the lower and upper troposphere) indicate CO enhancements in the upper troposphere (e.g., above 500 hPa) of 50 ppbv or more (relative to the minimum in the vertical profile). Interestingly, these enhancements often (but not always) appear without corresponding features in the aerosol extinction profile, possibly indicating 'rainout' of the aerosol. This feature of the CO profiles should be investigated in the revised manuscript and the potential role of deep convection should be addressed generally.

This is an important point and one mostly missed in the discussion manuscript. Based on the recommendations an analysis was conducted on each individual profile that extended through sufficient depth of the troposphere to identify enhancements in CO linked to deep convection. CO enhancements above the boundary layer were observed in 80% of profiles. However, only rarely was a similar increase seen in the aerosol products, indicating rainout as the referee suggests. This is consistent with the rBC coating thickness measurements. A summary of the changes to the supplement and manuscript to reflect this new topic area is provided below:

Method description: Sect S2, Supplement

Pollutant transport via deep convection: CO profiles with a sufficient vertical extent, of at least 5 km, were identified. The altitude of the CO minimum was identified and was used to determine a representative background concentration for the altitudes above and the altitude below. The altitude of the CO maximum above the minimum was identified and a representative CO maximum calculated from the concentration at that altitude, the altitude above and the altitude below. If the representative maximum CO concentration was greater than the representative minimum CO concentration plus a threshold of 40 ppb, transport via deep convection was identified. The rBC and σ_{sp} values were interrogated at the CO minimum and the altitudes of the maxima. If these were greater at the maximum altitude by 0.2 $\mu\text{g m}^{-3}$ (rBC) or 25 Mm^{-1} (σ_{sp}) than transport via deep convection for these aerosol properties was also identified.

Results: Table S3, Supplement

	All	EO	W1	W2	N1
CO Deep convection	81.1 (53)	100 (2)	77.8 (18)	79.3 (29)	100 (4)
Co-incident rBC increase	8.1 (37)	0 (2)	10.0 (10)	9.5 (21)	0.0 (4)

Co-incident σ_{sp} increase 2.7 (37) 0 (2) 0.0 (11) 4.8 (21) 0.0 (3)

Table S3. Percentage of sufficiently deep profiles showing evidence of CO transport via deep convection and co-incident increase in rBC and σ_{sp} . Bracketed values represent the number of sufficiently deep profiles.

Manuscript results, Sect 3.2:

There is evidence of moist convection delivering CO to altitudes above ~4 km but with significant wet scavenging of aerosol. In 81% of profiles with sufficient vertical coverage CO loadings increased by more than 40 ppb at altitude between ~4km to the top of the profile. Unlike the discrete signal from plumes, the enhancement was often 1-2 km deep. In contrast only 8% and 3% of the rBC and σ_{sp} profiles had a similar increase in signal co-incident with CO enhancements. This indicates significant removal of aerosol during convection to altitudes above ~4km across the atmosphere of Amazonas during the biomass burning season.

Manuscript discussion, Sect 4.1:

The shapes of pollutant vertical distributions are primarily controlled by meteorological conditions, in particular vertical convective motions and horizontal wind shear (Fig. 7). The former acts to mix pollutants released near the surface toward the mixing layer top, the altitude of which can be modulated by the latter, soil moisture and solar insolation. The difference in profile shape from west to east to north is primarily driven by contrasting mixed layer depths. Pollutant loadings remained relatively high above the mixing layer in residual layers, indicating wet removal is not significant at these altitudes. Large unmixed plumes perturbed the mixed and residual layers, although they contribute only 15% (E0), 11% (W1), 8% (W2), and 1% (N1) to the scattering only column AOD (calculated from the nephelometer, Sect. S2). Such plumes were seldom seen above 4 km, in contrast to previous observations in the region (Andreae et al., 2004), indicating the mass flux from large pyrocumulus detrainment into the upper troposphere (within the aircraft range) was not significant. The observed increases in CO concentrations above ~4km indicate vertical transport of mixing layer pollution into the free troposphere. The presence of co-incident increases in rBC or σ_{sp_dry} in less than 10% of these plumes indicates moist deep convection transports CO and presumably other gaseous and non-soluble components to altitudes greater than 4 km but efficiently removes aerosol from the air by wet scavenging. This is consistent with the decrease in rBC coating thickness at these altitudes in W1, W2 and N1 and also is similar to previous observations in Boreal Canada that showed preferential wet deposition of the largest and most coated particles (Taylor et al., 2014). As deep moist convection is not common in eastern regions (e.g. using TRMM rainfall as a proxy, Fig. 4a) the source of elevated and enhanced CO is unlikely to arise from the mixing layer in the east. It is possible the source is from CO aloft in the west which is recirculated in the persistent anti-cyclonic flow at 500 hPa (Fig. S2.f) as has previously been observed from satellite (MOPITT) CO observations by (Deeter et al., 2018). A long aging time is consistent with the larger rBC coatings observed aloft in E0.

Minor Revisions and Technical Corrections

page 5, line 17. Please provide reference for climatological winds over South America (e.g., Campetella, C. M., and Vera, C. S., *Geophys. Res. Lett.*, 29, 1826. <https://doi.org/10.1029/2002GL015451>, 2002).

Thank you for the reference, this has now been included:

‘In general, observed profiles of horizontal wind speed reflect those expected based on our understanding of synoptic flows over TSA (Campetella and Vera, 2002).’

p. 6, l. 26. Unclear if 80% refers to number of profiles where any or all of the considered pollutants indicated a pollutant residual layer

The sentence has been amended to make it clearer and now includes a cross-reference back to the originating table.

‘Over 70% of profiles included a pollutant residual layer of rBC, σ_{sp} and CO, even those in remote regions away from fresh emissions (Table S1).’

p. 7, l. 24. What about the east-west distribution of fires? The figure indicates many more fires in the eastern region. Is this typical?

The distribution is typical in that fire number is greatest in the east, although 2012 did feature more fires in the east than the 2008-2017 average. We decided against including a detailed climatology of fires and trends in the paper to keep it to a more readable length and focus the narrative.

p. 8, l. 1. Might stronger easterlies actually promote (rather than inhibit) the spread of fires?

If we were dealing with wildfires that may be true – however as most of the fires are managed by landowners, they tend to ignite when wind speed is low to enable greater control of the burn. This section has been rewritten based on the more detailed comments by reviewer 2.

p. 8, l. 17. Add 'significantly' before 'affected'. There must be some small effect, correct?

This has been amended – there is a small effect rather than no effect.

p. 8, l. 27. It seems surprising that CO at the surface decreases from W1 to W2 (from 340 to 220 ppbv) whereas CO₂ increases slightly (from 394 to 397 ppm). Does this suggest biogenic influence?

Yes, that is correct, this was noted on P9 L5 of the original manuscript. P9 L18 in updated manuscript.

p. 9, l. 18. If this statement ('Significantly, the shift ...') is based on Fig. 9, should the end of the sentence actually read '... the relative abundances of rBC and sigma_{sp_dry} *to* CO'?

Thanks to the referee for pointing this out. The sentence has been modified to read:

Significantly, the shift in meteorology between these two phases does not substantially impact the relative abundances of rBC, σ_{sp_dry} and CO to each other.

p. 9, l. 29. This particular paragraph ('The shapes of pollutant vertical distributions ...') seems largely qualitative and speculative. For example, sentences 5, 6, 7 and 9 in this paragraph draw conclusions without providing any quantitative evidence.

This has been revised (see the text from section 4.1) in the response to the general comment above.

p. 10, l. 28. Missing 'and' between 'phase' and 'plume'?

This section has been re-written following comments by reviewer 2.

p. 12, l. 14. SAMBBA was conducted in a year which was not considered a 'drought year' for the Amazon Basin. Widespread drought, such as occurred in 2010 and 2015, and may be increasing in frequency, results in different patterns of emissions (and meteorology) compared to non-drought years. Would the main findings of this paper be sensitive to the effects of drought? This would be an appropriate discussion for the Conclusion.

We thank the reviewer for a useful suggestion to widen the discussion. The following paragraph has been included in Discussion-Implications (Sect 4.2):

'Although the magnitude and bearing may differ, the fundamental drivers of the pollutant vertical distribution identified here will remain so in drought years which may be increasing in frequency (Jiménez-Muñoz et al., 2016). Dry convection may be more vigorous and the atmosphere more stable, deep convection less vigorous and aerosol scavenging reduced, fires more intense and fire hotspots located in different regions, but so long as model simulations well represent the fundamental drivers identified in this work then they ought to be able to replicate the resultant vertical distribution. This is a promising avenue for future research to predict the impacts in future years, following on from the study of Thornhill et al., (2018).'

We thank the anonymous reviewer for her/his thorough evaluation and constructive recommendations for improving this manuscript. Her/his comments (in blue) and our responses are listed below.

The authors present a vast body of very important data on the state of the atmosphere over Amazonia as evidenced in part by an incredible 168 page supplement with about 200 vertical profiles; each with many variables. It's been six years since the data was collected indicating that time for a very significant analysis effort occurred. There is a lot to think about. Caveats on my review: I'm not a meteorology expert and I only had time to read quickly thru the text and figures twice with only a glance at the supplement.

One point to raise is that there are two different important questions: 1) what is the state of the atmosphere? and 2) what causes it? The paper seems stronger on the first point, and information in regard to the first question is important even if the second one cannot be answered definitively.

My overall recommendation is to check the units (for BC especially) and figure captions carefully and to make sure a few angles of analysis (which I will list next) have been considered adequately. Most or all of the ideas on my list were probably considered by the authors, but since little or no mention is made, the reader can't be sure what has and has not been considered. Regarding the issues on my list, my expectation is that the authors can likely easily dismiss some, others may be sources of uncertainty in the pollutant profiles or driving forces, but yet others may yield insights. Most important is how these issues could impact the certainty about the actual pollutant structure.

Issues to address include:

1) Possible changes in the mass scattering coefficient with age (Akagi et al., 2012 in ACP).

See the comment below

2) The possibility of secondary organic or inorganic aerosol formation with aging.

The above two points relate to changes in aerosol composition and therefore optical properties with age. This is the focus of a recent paper by Morgan et al., (2019), which investigates aging of biomass burning aerosol in both the near and far field based on SAMBBA measurements. We note that there is no indication in this work of substantial increases in the mass scattering coefficient with age. Some near field changes in the mass scattering coefficient with age have previously been observed (Akagi et al., 2012). However, the work here is focussing on regional distributions and changes and individual fire plumes are not a significant component of the study and are unlikely to affect the results presented here.

3) The potential effects of aerosol concentration and ambient temperature on gas particle partitioning. Clues to these issues might possibly come from e.g., the coating thickness to BC ratio, neph-scattering to BC ratio, neph-scatter:SP2-scatter?, and O:C ratio of any AMS spectra collected or SPMS data that may have been even occasionally collected. These issues are addressed as part of a new section, which covers many of the line by line comments regarding the source of aerosol aloft. See (a.) below.

4) The SP2 is good for BC. Was there any info on BrC?

Unfortunately not – no multi wavelength absorption measurements were made using the aircraft.

5) Could dust or PBAP, e.g. super-micron aerosol have a regional role that should be put in perspective?

We examined the aerosol size distributions from the SMPS and GRIMM and also determined the Angstrom exponent at Red/Green to show that there was little contribution of dust or biological particles to the optical properties of aerosol in the column. We address this in the manuscript with the following figure and sentence in section 4.1:

'Consistent values of the scattering angstrom exponent at 700/550 nm within the boundary layer (Fig. S5) indicate a similar aerosol type throughout. These values were all above 1.5, typical of submicron biomass burning aerosol and indicating no significant regional role for super-micron dust or primary biological aerosols which have values closer to zero (Clarke et al., 2007; Russell et al., 2010). This is consistent with size distributions reported and from the aircraft (Darbyshire et al., in preparation).'

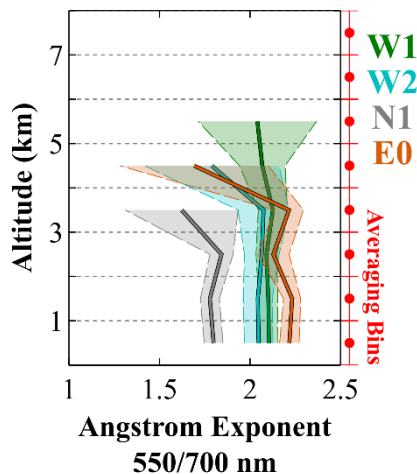


Fig S5. Median profiles of the scattering Angstrom exponent between 550 and 700 nm for each regime. Angstrom exponent calculated from nephelometer scattering coefficient measurements at these wavelengths. To reduce the noise the Angstrom exponent is only displayed here when the scattering coefficient at 550nm was greater than 10 Mm^{-1} . Lighter shading represents the median absolute deviation.

6) Was any use made of GOES data?

The resolution provided by MODIS data provides better spatial coverage than GOES and additional discussion of further satellite information was felt unnecessary in what is already a lengthy paper.

7) Briefly, how was “aerosol:CO” calculated and what are it’s units? Is aerosol:CO related to coating thickness and are there any insights at all from the AMS that was on board?

Aerosol:CO is used as a generic term to represent both scattering:CO and rBC:CO ratios which displayed similar behaviour. These enhancement ratios were calculated from plume integrated values above the local background (moving 5th percentile, i.e. grey ‘background’ in Fig. 3) and only when the two pollutants were correlated (Pearson’s $r > 0.5$). We are sorry that this was confusing and have added a sentence in section 3.4 of revised text to make this clear:

‘We use the term $\Delta\text{aerosol}:\Delta\text{CO}$ to describe both $\Delta\text{rBC}:\Delta\text{CO}$ and $\Delta\sigma_{\text{sp,dry}}:\Delta\text{CO}$ since these aerosol properties co-vary.’.

There was an AMS on-board, the vertical distributions from which will be presented in a forthcoming publication from this author, which presents detailed information on the aerosol composition and physical properties

Deep convection was mentioned by other reviewer and Yokelson et al., (2013, AMT) have a paper on the possibility of mixing (especially BL/FT) to change source ratios.

We thank the reviewer for the reference and note the point with the following added to section 3.4:

‘As these enhancement ratios are typically within the boundary layer, share a common source and in most cases are likely relatively fresh, it is unlikely they are biased by sudden changes in the composition of background air driving the observed gradient, as warned against in Yokelson et al. (2013).’

We reply to the issue of deep convection in response #1.

So in general, check all figures and units carefully and maybe do one more brainstorming run on the interpretation? A careful empirical summary of the observations without complete proof of why they occur is also useful.

We have now checked all the figures thoroughly.

Comments by line number:

Where the referee has made a number of comments on one subject, we have grouped the comments together and provide a combined response:

a) Cause of increasing aerosol:CO with altitude

We thank the reviewer for their detailed comments on this section, which have led to a significant re-working that has, we believe, improved the paper. The following comments are addressed in a re-written section which can be found below. Where comments are not directly addressed in this new text we have responded underneath that comment.

P1, L31-33: I'm not sure about this hypothesis as explained in detailed comments. Also MCE can be hard to measure away from the source (Yokelson et al 2013 AMT), especially in the free troposphere. BC may be a good independent indicator of "flaming induced plume rise" if/when that happens.

See below

P6, L18-21: It makes sense that the atmosphere is more often well-mixed in afternoon and we and others have seen that, but why would BC be well mixed but not CO if they are from the same source?

See below

P7, 4-5: Why are all pollutants not elevated in plumes? Variability, mixing, and other sources are mentioned and make sense especially for CO₂. But for CO, BC, and scattering; maybe the threshold is too high or plume signatures are erased by mixing?

This is the crux of what we are trying to understand, not only are the ratios of the aerosol properties to CO elevated in the averaged profiles, but also within the plumes. Mixing ought to mix out all the species equally, not some but not others, provided that their sources are all the same. We do not believe that we alias the ratios in the plumes as a result of the plume thresholds being set too high. We do show all of the profiles in the supplementary and there are no indications of enhanced values of variables sitting below the thresholds co-incident with plumes of other variables that exceed them. Unfortunately, because of the variability in the CO₂ concentration, mentioned by the reviewer, it was not possible to reliably retrieve MCE as a function of height which would have helped strengthen our hypothesis.

P8,L31: The aerosol emissions are usually higher at lower MCE in most studies, unless the authors have a good example in mind of the contrary?

The referee is correct that aerosol emissions do increase as MCE decreases and we have certainly observed this previously in laboratory and single plume studies. However, since we are comparing aerosol emissions to CO it may be that even though the aerosol emissions may increase as MCE decreases the proportional increase in CO is greater.

Seeing PM/CO (g/g/ or scat/ppm) larger at higher altitude could indicate PM evolution or that high MCE does not cause high plume rise? Other factors that influence plume rise, such as fuel consumption rate or atmospheric physics, might be more important?

See below

P10, L22:

In reality, it should be remembered that "plume injection altitude" is often not a fixed number, but an evolving mass-flux-vs-altitude, surface to plume top, distribution function (see Stocks et al (1996, JGR) SAFARI-92 paper).

P10, L25-34:

A number of things besides MCE could influence "aerosol"/CO. For example, secondary organic or inorganic aerosol, temperature, the increase in MSC with aging, and fire intensity as noted in Reid et al., (1998) where more intense fires had much higher EF-PM similar to the finding for wild vs prescribed fires in Liu et al. (2017).

In the new manuscript section below we discuss how for a given burn the fire intensity (as in heat released per unit length of fire front) is likely linked to the fire phase. This is simplified by the figure below:

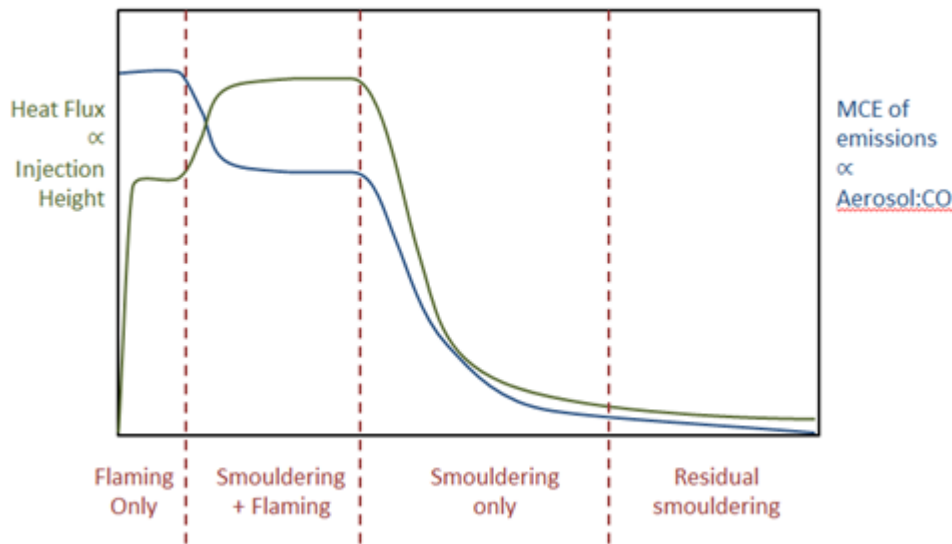


Figure. Idealised cartoon showing link between MCE and heat flux (y axis) and fire genesis (x axis) for a hypothetical single fire. (n.b. not included in manuscript)

As the reviewer points out, it is possible that this relationships fall down for very intense fires. Previous studies have shown that as an oxygen deficiency in a large flaming zone can reduce the combustion efficiency and therefore the emission of aerosol:CO would decline (Ward and Hardy, 1991). As far as we are aware there is limited study of this in the scientific literature after this study and therefore it is not possible to comment on how likely or frequent this phenomenon will be in tropical South America. We note that In the SCAR-B project referred to, Reid and Hobbs (1996, doi <https://doi.org/10.1029/98JD00159>) looked at how fire intensity controlled size parameters rather than EF-PM in Amazonian fires. As such, we recommend investigation into oxygen deprivation in active flame zones as an area for future research.

Fires tend to be more intense later in the day when the mixing layer is also deeper.

The referee makes a fair point. Under the assumption fires remain a similar size then these more intense fires could be characterised by a shift the upper limit of the y-axis in the above figure, meaning one would expect a greater injection height but also a higher integrated MCE. Therefore, assuming a similar fuel bed, one would expect to see the aerosol:CO increase with height as observed here. Note, that there would be a complicating factor given the (typical) strengthening of atmospheric stability in the daytime.

It's also possible the authors have the relation between MCE and EFPM reversed as noted above. I don't recall any of the cited works supporting the author's claim that aerosol/CO increases with MCE, but if they do, it might help to point out the specific figure or table where that evidence occurs?

In Figs S8 of the supplementary material, we show the EFaerosol vs MCE using data from Ferek et al., (1998) who measured PM4 and Yokelson et al., 2007 who measured PM10. In figure S9, we show values from Hodgson et al., (2018).

The phrase "combustion completeness phase plume injection height" is awkward and at a minimum, perhaps missing a word or dash to be clear? Further, "combustion completeness" is often used to describe the fraction of fuel consumed and Akagi et al (2011) actually argue that "phases" do not exist on real fires. The speculation is "OK" but it might make more sense to include a few candidate ideas about what drives the "aerosol"/CO rather than try to choose one based on the limited available evidence and resulting uncertainties.

See below

P11, L29-33: I think the description of fires is potentially misleading and other driving factors for aerosol/CO noted above deserve mentioning here. Small fires in the tropics in homogeneous fuels often are mostly a steady mix of emissions entrained into a column by flaming. Residual smoldering could play a role, but explicitly accounting for post-convection, residual-smoldering emissions would require separate EF and separate fuel consumption, which is not trivial to estimate. Estimates are available in JGR papers by Bertschi et al 2003 and Christian et al 2007, specifically for Brazil.

See below

I think the key here is to characterize the structure and the uncertainty in the structure, with a list of possible causes (examples above) being nice, but perhaps presented without choosing a preferred option, unless additional evidence can be presented also.

See revised text below

P12, L1-12: I kind of glazed over trying to follow this, so I'll just interject a detailed caution against assuming MCE and plume rise are positively correlated while freely admitting that they could be related at times. Converting fuel carbon to CO₂ releases more energy than converting it to CO, but buoyancy is related to air temperature and the rate of fuel consumption or CO₂ production. A fire burning with lower MCE could consume fuel at a higher rate in heavier fuels. Even for a fixed fuel loading, imagine a flame front at the beginning of a fire burning with an MCE near 1. As soon as it moves there is smoldering in its wake and MCE drops, but total CO₂ is increasing for a relevant-sized area because more fuel is burning. Air temperature above the fire peaks when CO₂ peaks, not MCE. That is what has been seen in most of our lab experiments or field work with towers or low flying helicopters. E.g. in Akagi et al 2012 an 81 ha fire had similar MCE at different heights in the convection column and the plume rise was dependent more or less on the instantaneous area burning. If you watch tropical fires, plume rise often peaks when flame fronts started on opposing edges converge, and then they annihilate. Secondly, even if the plume has more thermal energy, it may not rise higher ultimately. Plume rise depends on atmospheric stability, entrainment, the vertical profile of the winds, temperature, moisture (condensation can release energy), etc. The authors are in a good position to advise on the typical thermodynamics! Thirdly, flaming vs smoldering may impact the plume rise, but to check further maybe try some other F/S indicators like BC/CO that might work better or independently.

See the revised text below

Addition to Sect. 4.1 to address grouped comments:

'To try understand the relationship between $\Delta\text{aerosol}:\Delta\text{CO}$ and altitude it is necessary to examine multiple potential drivers. Substantial dry deposition could remove near surface aerosol and reduce the observed $\Delta\text{aerosol}:\Delta\text{CO}$. However, dry deposition fluxes reported in the literature (Ahlm et al., 2010) are small compared to the number concentrations observed (Darbyshire et al., in prep) so this is an unlikely cause. Increasing aerosol concentrations with altitude are typically attributed to partitioning of organic and inorganic species from lower volatility gas phase species into the particulate phase as temperatures decline and relative humidity increases (Heald et al., 2011; Morgan et al., 2010). A significant fraction of boundary layer organic aerosol in the regional haze will be secondary in nature following oxygenation and condensation of semi-volatile vapours during smoke plume evolution. During SAMBBA no net addition of organic mass was observed in the near or far field (Morgan et al., 2019), unlike previous studies (e.g. Akagi et al. (2012)). However, the contribution of further biomass burning secondary organic aerosol owing to increasing altitude is difficult to determine with the data available. There is an increase with altitude of the contribution from secondary inorganic aerosol with altitude in E0, but not W1, W2 or N1 (Darbyshire et al., in prep). This may account for the increase in rBC coating thickness with altitude (Fig. 10), but the decrease with altitude in W1, W2 and N1 indicates no significant addition of secondary organic or inorganic aerosol. Furthermore, as the approximate three-fold increase of involatile $\Delta\text{rBC}:\Delta\text{CO}$ with altitude is of the same magnitude to that of $\Delta\text{osp}:\Delta\text{CO}$, these secondary aerosol processes cannot explain our observations.

Plume $\Delta\text{aerosol}:\Delta\text{CO}$ enhancement ratios increase with altitude within the boundary layer, indicating that these plumes may be the source of the observed $\Delta\text{aerosol}:\Delta\text{CO}$ profiles in the regional haze. Past measurements (Fig. S8; Ferek et al. 1998; Yokelson et al. 2007) and those from SAMBBA (Fig. S9, Hodgson et al. 2018) indicate $\Delta\text{aerosol}:\Delta\text{CO}$ increases with MCE. We speculate the distribution may be driven by the relationship between MCE and fire intensity – and thus by extension, when normalising for meteorology, plume height (Lavoué et al., 2000). The peak heat flux (i.e. intensity) of an open landscape fire occurs when there is both a flame front, where flaming combustion occurs (high MCE), and zone of smouldering combustion (moderate MCE) in its wake. Once the flame front is extinct the fuel bed remains smouldering (moderate MCE) and although reduced, there may still be sufficient heat flux to generate a convective column to loft these emissions, albeit to intermediate altitudes. Once the heat flux is insufficient to generate any convective plumes emissions from residual smouldering (low MCE) are released at the surface. Together one would thus expect a continuum from higher altitude release of high + moderate MCE emissions to surface release of low MCE emissions, which could explain our $\Delta\text{aerosol}:\Delta\text{CO}$ observations. The evolution of plume height as a function of heat flux has previously been observed for savannah fires in South Africa (Stocks et al., 1996). Alternatively, there may be multiple convective cores for a given burn as seen elsewhere (Achtmeier et al., 2011). Where these are away from the primary flame front the MCE will be lower along with heat flux and therefore injection height. We recommend further fire-scale plume dynamics observations in tropical regions to test these speculations and acknowledge that many case studies would be required to account for the other impacts on 'real world' fires

plume rise (including stability, vertical profile of wind speed/direction, latent heat release, flame front annihilation, topography).

In addition to being related at the individual fire level, regionally there is a connection between MCE, average fire intensity and injection height. Fires in eastern Cerrado regions have a higher MCE (Hodgson et al., 2018), are more intense as illustrated by the FRP distributions in Fig. 5b and Gonzalez-Alonso et al. (2018), and have higher plume injection heights in spite of more stable conditions (Fig. S10, Gonzalez-Alonso et al. 2018; Marengo et al. 2016).

b) Scaling Factors

P3, L2: At least four papers, including two from the FAAM group by Hodgson et al and Reddington et al seem to prove that missing fires is the biggest reason for emissions errors. The two non-FAAM papers are here:

<https://agupubs.onlinelibrary.wiley.com/doi/epdf/10.1029/2018GL078679> <https://www.atmos-chem-phys.net/11/6787/2011/acp-11-6787-2011.html> (See Table 1 on fire detection).

P12, L3: The Reddington et al scaling factor mentioned earlier in this paper and the other papers I noted above in this review overwhelmingly suggest that missing fires is the problem rather than, e.g., a factor of five on emission factors.

For reference, here is P3, L2:

“Furthermore, to ensure consistency with remotely sensed AOD measurements, emissions are typically up-scaled by a factor of two to five (Johnson et al., 2016; Reddington et al., 2016). There is no established physical understanding of this requirement, with deficiencies in both emissions inventories and model process posited. As such, scaling factors represent a key uncertainty in model treatment of biomass burning across the globe”

And P12, L3:

“Past satellite observations have suggested residual smouldering is a large source of CO and not fully captured by emissions inventories (Deeter et al., 2016; Pechony et al., 2013), which may be based on inappropriate emissions factors or arise from small fires outside the detection limit of active fire detection schemes (Giglio et al., 2016; Schroeder et al., 2008).”

Scaling factors to reconcile model and satellite or AERONET AOD are likely required because of two components, deficiencies in emissions inventories and deficiencies in model treatment. The reviewer contends that the deficiencies in emission inventories are not caused by the emissions factors, rather a misrepresentation of burned area owing to undetected smaller fires. Such a misrepresentation is observed in Florida by Nowell et al., 2018, however it is not possible to determine if this would successfully reconcile modelled and measured AOD. This is because Florida is a humid environment and water uptake would likely increase the AOD. As the amount of water uptake is itself an uncertain process, it is not currently possible to derive a ‘dry’ AOD with which to compare ‘dry’ model output. Therefore one cannot definitively say that *‘missing fires is the biggest reason for emissions errors’*. Even if this were the case for Florida, it is not clear that the discrepancy between real and observed burned area would be the higher, lower or the same in tropical South America, where burning practices differ.

However, there is evidence from SAMBBA which does point towards the problems with emissions being from missing fires - the emissions factors in Hodgson et al., (2018) are consistent with those previously reported for the region, whilst Reddington et al., 2016 and 2018 show that using an emission inventory which includes small fires (FINN1) compared to one which doesn’t (GFED3) does reduce the scaling factor required to 1.5. Again it is not possible to evaluate the overall effect because of the complicating factor of humidity, as explored by Johnson et al., (2016), Reddington et al., (2018) and a forthcoming paper from this author. Therefore, it cannot be discounted that emission factors may still not best represent emissions from biomass burning in tropical South America.

This is summarised by the following amendments to the manuscript:

Update to P3, L2 Original manuscript. (P2, L34 updated manuscript)

“Furthermore, to ensure consistency with remotely sensed AOD measurements, aerosol emissions are typically up-scaled by a factor of two to five (Johnson et al., 2016; Reddington et al., 2016). This represents a key uncertainty in model treatment of biomass burning aerosol, with deficiencies in both emissions inventories and model process posited.”

Sect 4.1

‘Consistent values of the scattering angstrom exponent at 700/550 nm within the boundary layer (Fig. S5) indicate a similar aerosol type throughout. These values were all above 1.5, typical of submicron biomass burning aerosol and indicating no significant regional role for super-micron dust or primary biological aerosols which have values closer to zero (Clarke et al., 2007; Russell et al., 2010). This is consistent with size distributions reported and from the aircraft

(Darbyshire et al., in preparation). This also indicates that model scaling factors to match remotely sensed AOD are not significantly biased by non-biomass burning aerosol. The observed increase in humidity with altitude in the boundary layer (Fig. 7C) will likely have a significant impact on AOD and therefore the required scaling factor as it remains an uncertain model process (Johnson et al., 2016; Reddington et al., 2018). We note that findings from SAMBBA suggest that omission of burned area from small undetected fires is the most significant source of inventory under-representation of aerosol emissions (Hodgson et al., 2018; Reddington et al., 2016b), as in other burning regions (Nowell et al., 2018).'

Update to P12, L3 original manuscript. Moved to section 4.2 (P13, L17 in updated manuscript)

We speculate the Δ aerosol: Δ CO profile may be driven by a coupling between combustion efficiency and plume dynamics. If confirmed by further enquiry then future modelling studies will have to consider how best to represent the phenomenon. Replicating the gradient Δ aerosol: Δ CO may be particularly important for model simulations which draw results based on a realistic vertical distribution. For example aerosol-cloud and aerosol-radiation interactions, or surface air quality simulations. Emissions factors for residual smouldering combustion in the Amazon have been collected (Bertschi et al., 2003; Christian et al., 2007), and could be used to test predictions from novel (and non-trivial) model set-ups against surface and satellite observations. Past satellite observations have suggested residual smouldering is a large source of CO and not fully captured by emissions inventories (Deeter et al., 2016; Pechony et al., 2013).

c) Units

P1, L22: First place I noticed where units on BC may be off?

P6, L24: delete "c" in "cm-3"?

P8, L20-22: Again here and throughout: Am I understanding the units right? 120 Mm-1 would be about 40 ug/m3 of PM2.5 – but the rBC of 1.7 ug/cm3 would be about 1.7 E6 ug/m3. In Fig S3, the rBC peaks about 1.5 ng/sm3, which is 9 orders of magnitude lower? Then in the 198 profiles the BC is about 1 ug/m3, which is different again, but the ug/m3 units seems to make the most sense throughout.

The referee is correct and many thanks for identifying the error. After tracking, the mistake arose from the units previously being in $\mu\text{g sm}^{-3}$, where 's' inferred standard atmosphere – this was advised against in a co-author review and inexplicably the 's' was changed to a 'c' rather than removing completely. Apologies for the confusion caused.

d) Remaining comments

P1, 16: Reduced uncertainty in processes may be less important than just presenting the wealth of vertical profile info.

We agree that the paper itself does not reduce uncertainty of impacts on weather and climate so we have rewritten this sentence:

We examine processes driving the vertical distribution of biomass burning pollution following an integrated analysis of over 200 pollutant and meteorological profiles measured in-situ during the South American Biomass Burning Analysis (SAMBBA) field experiment. This study will aid future work examining the impact of biomass burning on weather, climate and air quality

P1, L23: Can you really measure surface CO from a plane? Here and elsewhere, maybe "surface" should be defined as an AGL range?

The text has been amended to be consistent with language elsewhere in the manuscript via use of the term 'near surface'.

P1, L26: "Optically thin" or "thin layers of pollutants"?

In most cases, the plumes above the mixing layer are both altitudinally thin and therefore when integrating the extinction coefficient to derive the AOD, also optically thin when compared to those measured in the boundary layer. However as we don't consider the extinction coefficient in this manuscript, and to avoid the confusion you have identified, we have amended the sentence to simply read 'Thin layers above the mixing layer....'.

P2, L11: Clarify what is meant by haze lifetime? Average lifetime of a smoke plume from a typical contributing fire?

P2,L11 for reference:

"These uncertainties are pronounced in tropical South America (TSA), one of the largest global biomass burning sources. Aerosol accumulates within the convective boundary layer forming a regional haze with a ~~lifetime of 4-5 days~~ (Edwards et al., 2006) that can cover up to 6 million km² (Prins et al., 1998) and reach aerosol optical depths (AOD) of up to 4 in the mid-visible during particularly polluted years (Artaxo et al., 2013)."

I have removed this section of the sentence to avoid confusion as the 4-5 days figure relates only to aerosol and is estimated based on the method and figure 7 in Edwards et al, (2006) and will be uncertain.

P4, L9: No mention is made of the AMS. How is “aerosol” measured and what are its units?

In the linked reference (Trembath, 2013) there is an analysis of the Rosemount inlet transmission efficiency for different particle sizes. The number and mass size distribution of biomass burning aerosol is dominated by submicron aerosols, and there is high particle transmission efficiency in this region (see Trembath, 2013). The SP2 is a sub-micron instrument. The AMS is not used in this analysis.

P4, L20: Which profiles, from the plane or the dropsondes or both?

Both, the sentence has been amended to include ‘Individual profiles from the aircraft and dropsondes were averaged...’

P5, L6 and L28: Is this a local maximum in wind speed not counting the free troposphere?

L6, refers to the trade wind inversion marking the border between the PBL and free troposphere. L28 refers to our observation which show that multiple wind speed maxima were observed through the column and one of these would typically be co-located with the top of the mixing layer. This has been clarified in the text with the following sentence:

At L6

Typically, a maximum in horizontal wind speed of variable magnitude and extent is present above the entrainment zone and referred to as the trade wind inversion.

At L28

A wind speed maximum is generally observed above or collocated with the mixed layer top (e.g. Fig. 3). At times this was the trade wind inversion but typically it was not.

P5, L10 and L27: Does “positive” wind shear mean an increase in headwind experienced by the aircraft, a decrease or increase in prevailing winds, a wind direction change, or something else?

We thank the referee for pointing out that this wasn’t clear in our original manuscript. This has been amended in the supplementary material (P3,L31) to include an explicit explanation:

‘Positive wind shear refers to an increase in horizontal wind speed with increasing altitude.’

Thanks to the referee for pointing this out since it has also raised an omission that we did not include information regarding the wind measurements in the methodology. Hence the following sentence has been added to the methodology section.

‘A five hole radome-mounted turbulence probe at the aircraft nose provides measurement of airflow relative to the aircraft, thus allowing calculation of wind vectors when combined with a GPS inertial navigation unit (Petersen and Renfrew, 2009).’

P7, L8&10: “lifted” > “lifting”

Amended

P7, L27: “Whist” > “Whilst”

Amended

P7, L30: AOD seems to peak downwind of the area of maximum fires, which is “OK”. This sentence may be better as “an enhancement in AOD of 0.1,” but this seems to be a very small AOD enhancement compared to the AOD of 4.0 mentioned earlier

We thank the referee for this comment. The AOD of 4 referred to earlier was an example of the extreme loadings possible as part of the introductory contextualisation. The average AOD map presented in Fig. 5a provides a more accurate representation of a typical year. The sentence has been amended based on this recommendation, from: “Aerosol accumulates within the convective boundary layer forming a regional haze with a lifetime of 4-5 days (Edwards et al.,2006) that can cover up to 6 million km² (Prins et al., 1998) and reach aerosol optical depths (AOD) of up to 4 in the midvisible during particularly polluted years (Artaxo et al., 2013).”

To:

“Aerosol accumulates within the convective boundary layer forming a regional haze that can cover up to 6 million km² (Prins et al., 1998) and whilst weekly averaged aerosol optical depths (AOD) are typically 0.75-1 in the mid visible, they can reach 4 in extremely polluted years associated with drought (Artaxo et al., 2013).”

P7, L29-32: Most of the fires are set by farmers, ranchers, or loggers, and they like to burn at low RH and wind speed to get good fuel consumption, but with reasonable ease of control/less chance of escape. An increase in either RH or wind speed means less suitable conditions for initiating burning. However, also, increased cloud cover can reduce fire detection efficiency.

The referee makes some very valuable points. The section has been amended to:

‘We speculate conditions were not optimal for human ignited fires, as a rise in relative humidity and increase in wind speed makes ignition and control more difficult, whilst decreasing the fuel consumption. The increase in cloud cover may have also reduced fire detection efficiency.’

P8, L11: Does E0 cover Sep 14 – Oct 5?

The referee makes a fair point. It is not clear that E0 and N1 do not represent the whole period. The bullet points have been amended to account for this.

P8, L18: How is “absolute aerosol burden” defined and quantified?

Aerosol burden was the collective term employed to collectively describe both the rBC mass loading and scattering coefficient. However we acknowledge this may be confusing and misinterpreted as solely mass. As such all instances of ‘burden’ have been changed to ‘abundance’.

The use of ‘absolute’ is perhaps confusing and has been removed from the sentence.

P8, L23: “surface” should be more precisely defined since we observed some instances of very high, reasonably widespread CO₂ (>5ppm) on the ground in the Amazon. P8, L24: “unknown” or is e.g. “variable” more appropriate? See comment for P1, L23.

P8, L26-28: Is 397 ppm the surface layer CO₂ in W1 and W2? Andreae et al 1988 found CO₂ at low altitudes really dependent on time of day (PS vs respiration).

We agree that this sentence is confusing – it has now been amended to:

‘CO₂ mixing ratios are also greatest at the near surface: 402 ppm in E0, 393 ppm in N1, 394 ppm in W1 and 397 ppm in W2 representing a particularly prominent enhancement.’

Fig 8 shows the difference between the morning and afternoon for CO₂ averaged across all flights. It is greater near the surface in the morning (respiration) than afternoon (photosynthesis). The role of biogenics on the CO₂ profiles is further discussed in P9 L18 in the updated manuscript.

P8, L31: How is the “aerosol burden” calculated? E.g. is there an inferred mass?

See comment for P8, L18.

P9, L2: Again excess aerosol should have units or some more precise explanation?

See response 7).

P9, L8-10: Here the authors do note a possible role of temperature and aging on coating thickness, which is great. Perhaps temperature and smoke age deserve broader attention?

See response 3).

P9, L11: Why the difference in coating thickness above the BL for different regimes? Do these regimes have a different concentration gradient above the BL so that evaporation due to a concentration decrease happens only in some cases?

The cause is more likely scavenging during wet convection. This point is made in response to the main recommendation of reviewer #1 – for more details see that response.

P9, L19: “influence” may be better than “determine”?

Amended

P9, L28: “combustion processes” may be better than “emissions”?

Amended

P10, L1: sentence may need one more word? E.g. “they” before “contribute”?

Amended

P10, L2-3: is the relative frequency of deep convection different from Andreae et al 2004 maybe because different geographic regions were sampled?

P10, L2-3 for reference:

“Such plumes were seldom seen above 4 km, in contrast to previous observations in the region (Andreae et al., 2004), indicating the mass flux from large pyrocumulus detrainment into the upper troposphere (within the aircraft range) was not significant.”

Many of the pictorial examples given in the supplementary material of Andreae et al., 2004 seem to be within or close to our sample region in Rondonia or south of Manaus. Their location tags are slightly at odds with the flight track presented in Andreae et al., 2004 Fig. 1, but consistent with another plot of the LBA-SMOCC flight tracks in Almedia and dos Santos (2007, <http://dx.doi.org/10.1590/S0102-77862007000300004>). So in summary, based on visual observations only, the relative frequency of pyrocumulus was lower during SAMBBA in approximately the same sample area.

P10,L2,L3 has now been slightly amended to read:

“Such plumes were seldom seen above 4 km, in contrast to previous observations in approximately the same sample region (Supplementary material in Andreae et al., 2004), indicating the mass flux from large pyrocumulus detrainment into the upper troposphere (within the aircraft range) was not significant”

P10, L6-7: Saying pollution persisted “above” the mixed layer seems potentially inconsistent with the earlier text about clean conditions at altitude.

This section has been re-written in response to reviewer #1 – for more details see that response.

P10, L9: “they exhibit”?

Amended

P10, L13: “eats” should be “east”?

Amended

P10,L13-14: “above the lifting”?

Amended

P11, L1: Maybe “remote sensing” is too broad since MOPITT a-priori profiles, and perhaps others, are flat over the whole column?

None of the studies referred to in this section are from MOPITT retrievals.

P11. L8: replace “This study” with explicit reference to a paper?

Amended

P11, L14-28: Good points.

P12, L29-31: Christian et al 2003 (JGR), Akagi et al., (2011), Andreae & Merlet 2001, and many other papers do already recommend higher BC/OC, BC/CO, etc for savannah vs forest.

This is correct and the Hodgson et al., results are consistent with these findings. The main thrust of this point was to recommend assessment of how the significant east/west difference in pollutants propagates to climate impacts via targeted modelling studies. As such the sentence has been amended:

‘Cerrado burns in the east are more flaming, whilst those in the west, of primary/secondary forest and pasture land are more smouldering (Akagi et al., 2011; Andreae and Merlet, 2001; Hodgson et al., 2018). The regional contrast in biomass burning emissions resulting from these different fire types clearly merits future investigation in modelling studies to assess potential impacts on aerosol optical properties and their radiative effects.’

P13, L10: Is this obvious from satellites or the average meteorology?

The prevailing low level easterlies are clearly identified in ECMWF reanalysis.

Figure comments

Figure 1. Its hard to tell the difference between high altitude surveys and straight level runs, make one orange or yellow or green?

We thank the reviewer for this suggestion. As Orange would clash with the box for 'East' and green with the LCSS land cover map, we have instead used a brown colour to ensure all features are distinguishable.

Fig 2. Should there be typical altitude values (and units) on the vertical axis?

We thank the reviewer for this suggestion. However we are of the opinion that additional altitude markers on the vertical axis will act to complicate the schematic. This is primarily because the vertical axis is not linear, but also because more precise altitudes from the layers we have observations for are provided in the accompanying legend.

Fig 3. There are two BB plumes identified on the rBC trace. There are no BB plumes identified on the CO trace, and only one each on CO₂ and Bscat. It seems like there should be two on all traces if they really are BB. Also, I find it hard to locate/digest the structure and plume features that are explained in the upper right boxes.

We thank the reviewer for this comment as it illustrates the figure explanation was not adequate. A biomass burning plume was identified if any of the species passed the identified threshold (dot-dashed line). The red trace illustrates this for each species, so for example at ~650 hPa the algorithm detects a plume for rBC and the scattering coefficient, but not for CO or CO₂. The intention was to explicitly illustrate this by having the species with an identified plume in red whilst those without remaining in black. As one or more of the species exceeded the threshold, our scheme defines this example as a biomass burning plume. It is worth noting here that if CO is well correlated (Pearson's $r > 0.5$) – it is in this particular example – then the plume will be included in the scatter plot in Fig 9. C-D.

The figure label has been amended to include:

Note, the red outline of a biomass burning plume is only present if it was identified for that specific species. For example at 65 kPa rBC and σ_{sp_dry} pass the identification threshold (grey dot-dash line) but CO and CO₂ do not.

Fig 4. How can the difference for soil moisture be in tens when the absolute values are fractional?

We thank the reviewer for pointing out this error. The values were scaled by a factor of 1000 to improve the figure appearance, but no explanation of this was provided. The text on the colour bar has now been altered to: "P1- P2 (x 1e3)"

Fig 5. Maybe label FC as "FC/100" also in Fig 6?

We thank the reviewer for this suggestion and have updated the figures accordingly.

Interesting that the AOD max is downwind of the detected fire max. This could be transport, but also different detection efficiency, higher PM emissions from more smoldering in forest fires.

We agree with the reviewer and that it is likely a mix of the suggested factors. We refrain from a detailed discussion of this in the text to ensure the manuscript remains focused on the vertical distribution.

AODs up to 4 that are mentioned in the text relate to this figure how?

The AOD of 4 referred to was an example of the extreme loadings possible as part of the introductory contextualisation. This sentence has been slightly amended for clarity from:

"Aerosol accumulates within the convective boundary layer forming a regional haze with a lifetime of 4-5 days (Edwards et al., 2006) that can cover up to 6 million km² (Prins et al., 1998) and reach aerosol optical depths (AOD) of up to 4 in the midvisible during particularly polluted years (Artaxo et al., 2013)."

To:

"Aerosol accumulates within the convective boundary layer forming a regional haze that can cover up to 6 million km² (Prins et al., 1998) and whilst weekly averaged aerosol optical depths (AOD) are typically 0.75-1 in the mid visible, they can reach 4 in extremely polluted years associated with drought (Artaxo et al., 2013)."

Fig 6. Would dates or definitions of dry and dry-wet transition seasons be useful? Yokelson et al., (2007) saw a huge burst in burning during a few days with the lowest RH.

We thank the reviewer for this suggestion and have updated the figure captions to include the dates. Whilst an analysis of fire activity with factors such as RH would be interesting, it is outside the scope of this paper and therefore we refrain from including it.

Fig 7. The figure doesn't appear to have any "pollutant" info that is referred to in the caption. Do you mean "meteorological"?

We thank the reviewer for pointing out this error. The caption should indeed read 'meteorological' and has thus been amended.

Fig 8. Check units on rBC. Could the CO and aerosol shapes be different partly because CO may be more impacted by non-fire (urban) sources that inject lower than fires?

rBC units altered to be consistent with rest of manuscript. Changed from $\mu\text{g sm}^{-3}$ to $\mu\text{g m}^{-3}$ ('s' had previously stood for 'standard').

We thank the reviewer for the suggestion and it has led us to more explicitly discount CO from urban sources as significant as the emissions hotspots are a significant distance away from our flight region and emissions are an approximately an order of magnitude lower than for biomass burning (see figure below, which has been added to the supplement). There was no influence on the average profiles from elevated CO at aircraft take-off/landing as these sections of individual profiles were removed. This is discussed in Sect. 4.1 of the manuscript.

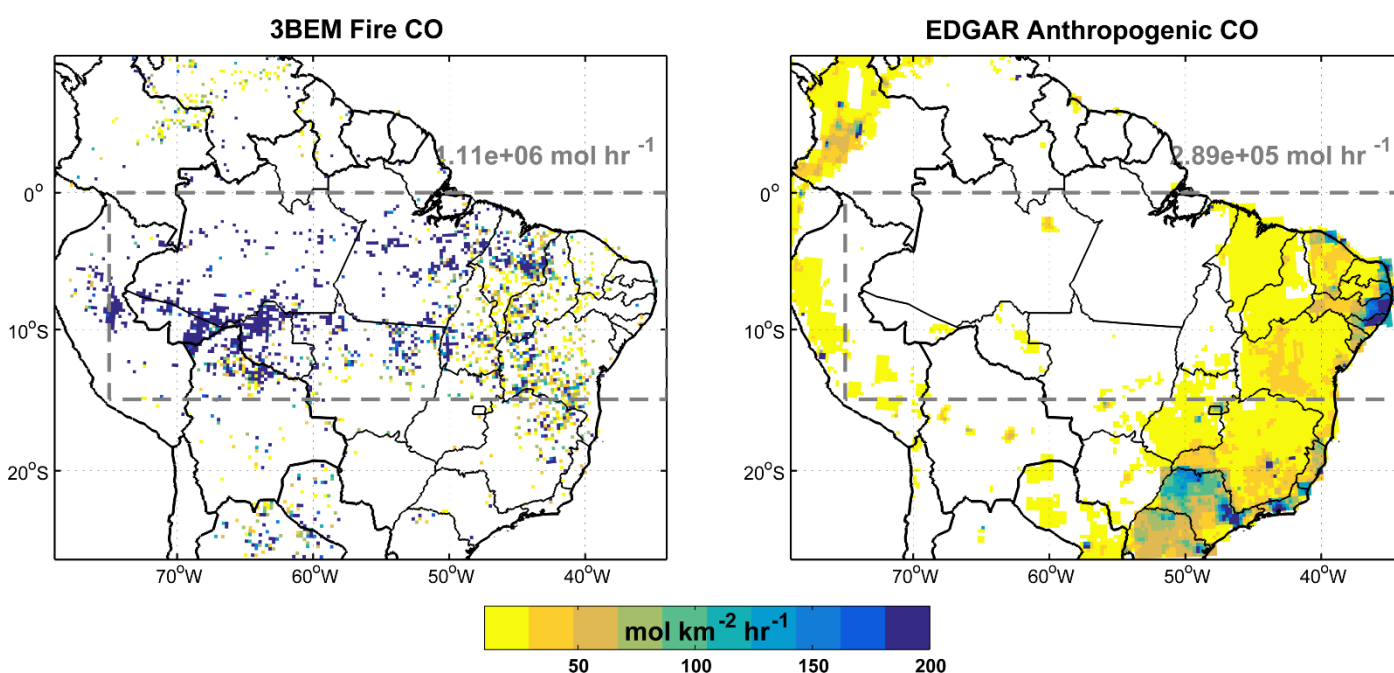


Fig S9. CO emissions on 20th September 2012 from biomass burning (left) and anthropogenic sources (right). Fire CO emissions are derived from the Brazilian Biomass Burning Emission Model (3BEM). Anthropogenic CO emissions are derived from the Emissions Database for Global Atmosphere Research (EDGAR) version 4.0 2005. Both emissions maps were generated using PREP-CHEM-SRC v1.4 as described in (Archer-Nicholls et al., (2015)). The dashed grey box represents the flight area and near-surface air mass history. The sum of the emissions flux within this area is 1.11 Mmol hr^{-1} for fire CO and 0.29 Mmol hr^{-1} for anthropogenic CO.

Fig 9. Needs units. More?

We thank the reviewer for bringing the lack of units to our attention. These have now been added to the figure. Unfortunately we do not understand what the "More?" comment refers to and seek clarification if the reviewer feels it important.

Fig 10. Any way to relate coating thickness to age, or size, or Bscat/BC or T, where the latter two might be related to gas to particle partitioning?

Coating thickness is related to deep convection and wet scavenging (Sect 4.1) and briefly, partitioning (Sect 4.1). Regarding aging, please refer to the comment 2 on page 1.

Fig 11. This is kind of overwhelming. Maybe help the reader with a few sentences about what it is trying to show.

We thank the reviewer for the comment and agree it is a busy figure. We have made minor alterations to the plot in an attempt to make it clearer. In addition we have re-written the figure caption:

Figure 11. Summary of the west to east regional gradient in equivalent potential temperature (i), relative humidity (ii), horizontal wind speed (iii) and aerosol extinction (iv), based on an east to west transect (Panels B) on the 27th September 2012 (flight b743). The thermodynamic curtains (B.i-iii) are derived from linearly interpolated aircraft (grey

line) and dropsonde (grey dots) profiles. The curtain of the lidar extinction coefficient at 532 nm (B.iv) is reproduced from Marenco et al., (2016) scaled from 355nm using a scaling factor of 0.57 following Marenco et al., (2014). The regional gradient is emphasised by the side panels which show the median thermodynamic and aerosol extinction profiles for all western (A) and eastern flights (C). In panels (i) to (ii) the observed thermodynamic parameters are compared to ECMWF reanalysis data extracted along the flight paths. In panel (iv) the in-situ aerosol scattering coefficient at 550 nm is compared to the lidar extinction coefficient averaged over a similar region (Fig. S6; n.b. both measurements are reported at ambient temperature and pressure).

Some minor alterations have been made to the paragraph discussing the figure in the text:

A transect flight from east to west (Fig. 11) captures and summarises the meteorological drivers of the regional contrast in pollutant vertical distribution. A declining mixing layer depth from east to west is evident from the reduction in altitude of the sharp gradient (i.e. the entrainment zone) of θ_e from ~ 3 km in the east to ~ 1.5 km in the west. Above the mixed layer top relative humidity increases, especially so above the lifting condensation level where the high humidity distinguishes the cloud convective layer. This layer is deeper in the west, associated with deeper moist convection. A wind speed maximum is present at 5-6 km, coincident with the entrainment zone. Together, this structure can explain the lidar derived extinction coefficient distribution (first published by Marenco et al., 2016). Aerosol is capped below the first wind speed jet, well mixed within the mixed layer and features a maximum at ~ 1.5 -2 km. Visible plumes at $\sim 59^\circ$ W and $\sim 52.5^\circ$ W lie at injection heights typical of those observed in the in situ profile data. The similarity of the in situ σ_{sp_amb} and lidar extinction coefficient profile shapes at the regional (Fig. 11) and local (Fig. S7) scales engenders confidence in the representativity of both datasets. Disparity in the absolute magnitudes is primarily explainable by differences in the sampling coverage (Fig. S8).