Comments on changes to Characterising Brazilian biomass burning emissions using WRF-Chem with MOSAIC sectional aerosol

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General Comments

Due to the large number of reviewers for this paper, there have been many changes to the text and figures in this paper. The discussion paper also formed part of S. Archer-Nicholls' thesis, and minor corrections noted by the external examiner have also been integrated into the changes. Finally, as measurements from the SAMBBA campaign are still undergoing quality control, some figures have been redrawn accordingly. This document will serve to highlight the line-by-line changes in the latex-diff file, justifying why changes have been made.

Line-by-line changes

Abstract

Pg2., lines 27-28

Values of single scattering albedo edited to reflect changes to aircraft data reported in Figure 9.

Section 1 Introduction

Pg2., Lines 44-45

References recommended by reviewer #2, comment #1 added.

Pg3., Line 61

Reference to Zhang et al., 2014. Added as recommended by reviewer #2, comment #2.

Pg3., Lines 71-85

This paragraph has been expanded and restructured to incorporate references and give credence to use of specified injection heights, from reviewer #2, comment #3.

Pg4., Line 101.

Noted that the parameters of the plume-rise parameterisation can be impossible to measure, after reviewer #2, comment #3.

Pg4., lines 106-110

Changes made to reflect MISR plume heights over South America, in response to reviewer #3, comment #4.

Pg4, line 112

Reference to Colarco et al., 2004 included after reviewer #2, comment #3.

Section 2 Model, emissions and the plume-rise parameterisation description

Pg5., lines 150-161

Paragraph has been edited to incorporate discussion of the modal aerosol mechanism used in WRF-Chem, after Reviewer #5, comment #1.

Pg6., lines 163-189

There have been numerous deletions I these paragraphs to condense this section, after recommendations to condense discussion by Reviewer #2, comment #5.

Pg6., lines 176-177

The reasons for not using the VBS have been edited, based on the comment by Reviewer #3, comment #1.

Pg.6, lines 190-192

It is made explicitly clear here that the Maxwell-Garnet mixing rule is used in this study, in response to J. Reid comments #2 and reviewer #5 comment #2

Pg7., lines 201-209

These lines have been deleted to condense this section, after recommendations to condense discussion by Reviewer #2, comment #5. The discussion in this section was largely repeated in the Analysis, Discussion and Conclusions section of the paper, and so considered unnecessary here.

Pg7., lines 220-229

Paragraph rephrased and shortened after recommendation to condense discussion by Reviewer #2, comment #5.

Pg8., lines 246-250

Paragraph edited to incorporate reference of Zhang et al., 2014 as recommended by Reviewer #2, comment #2.

Pg8. line 253 - Pg9., line 293

Numerous edits and deletions to shorten text after recommendation to condense discussion by Reviewer #2, comment #5.

Section 3 Model and emission product developments

Pg9., line 295 – Pg10., line 304

Paragraph rephrased and shortened after recommendation to condense discussion by Reviewer #2, comment #5.

Pg10., lines 295 – 321

Minor deletions and rephrasings to make discussion clearer and shorter after Reviewer #2, comment #5.

Pg10, lines 330-332

Reference to Peterson et al., 2014 added after Reviewer #2, comment #4.

Pg12., lines 371-385

Minor deletions and rephrasings to make discussion clearer and shorter after Reviewer #2, comment #5.

Pg12., lines 393-395

Paragraph rephrased to incorporate reference to Reid et al., 1998, after comment #3 by J. Reid.

Pg13., line 430 – Pg14., 457

Large section of text on the MACC-II model moved to an Appendix, in order to make it quicker for the reader to reach the novel parts of the paper, after Reviewer #2, comment #5.

Section 4 Campaign description

Pg15., line 477

Correcting previous error in manuscript – resolution should be 1x1 degree.

Pg16., lines 515-518

Unnecessary lines removed to clarify meaning based on recommendations and edits by coauthors and external examiner. In addition, reference to personal communication with J. Trembath removed due to him being a co-author of the paper.

Pg16., lines 540-542

Unnecessary lines removed based on recommendations by coauthors and external examiner. The location of the flights can be seen in Figure 7.

Pg17., lines 548-559

Paragraph expanded to include more rigorous definition of the boundary layer and comparisons with Fisch et al., 2004, after Reviewer #1 by comment #4.

Pg17., lines 560-570

Paragraph edited to clarify meteorology used to drive the MACC-II runs, after Reviewer #1, comment #3.

Pg18., lines 590-591

This line was removed after a recommendation by external examiner, as the description of Figure 2 is in the Figure caption and does not need to be repeated here.

Pg18., lines 593-606

This paragraph has been edited to incorporate changes to the boundary layer discussion after Reviewer #1, comment #4 and include the reference to South American injection heights after reviewer #3, comment #4. Following these changes, unnecessary lines were removed to improve clarity.

Pg19., lines 619-625

Paragraph edited and condensed, with unnecessary lines removed, to improve clarity on when and why nudging was used/not-used after Reviewer #2, comment #6.

Section 4 Results and Analysis

Pg.20, line 665

Explicit reference to the lowest temperature inversion added in response to Reviewer #1, comment #6.

Pg20, lines 678-682

Paragraph edited to incorporate discussion of importance of emissions as noted by Reviewer #2, Comment #7.

P21., lines 690-694

Discussion of statistics added after Reviewer #3, comment #5.

Pg22., lines 738-742

These lines were removed after a recommendation by external examiner, as the description of Figure 8 is in the Figure caption and does not need to be repeated here.

Pg23., lines 830-831

Values of measured single scattering albedo edited to reflect new flight data used in Figure 9.

Pg.25, lines 841-844

Paragraph expanded here in incorporate some detail deleted from Section 2, after recommendation to condense discussion by Reviewer #2, comment #5.

Pg.25, line 845

Note that Figure 10 now shows number and volume distribution, after Reviewer J. Reid, comment #6.

Pg.25, line 858

Error corrected, the measured CCN concentrations are between 0.135 and 0.154% supersaturation, not 0.14%.

Section 6 Conclusions

Throughout this section, effort has been made to condense the discussion and remove unnecessary lines, after Reviewer #2, comment #10.

Pg.27, lines 923-296

These lines have been edited in response to Reviewer #1, comment #8 on the difficulties of modifying the temperature inversion without the availability of hindcast data.

Pg.28, lines 952-255

Notes have been added here that there is ongoing work being conducted to incorporate the VBS for these SAMBBA runs, which be compared with more in-depth analysis of the compositional data from the SAMBBA flight campaign, in response to Reviewer #5, comment #3.

Tables

The tables have been reordered, in response to Reviewer #2, comment #3.

Table 4.

The caption has been condensed to make meaning clearer and remove text that is repeated in the main paper. In addition, an added column with the temporal resolution of the instruments has been added in response to Reviewer #2, comment #11.

Table 6.,

This table of statistical comparison between the model runs and MODIS data has been added in response to Reviewer #3, comment #5.

Figures

Figure 2.

This figure has been redrawn to make it easier to read, with scale revised and scientific notation used for the units.

Figure 3.

This figure has been redrawn with a new colour scheme in response to Reviewer #1, comment #5.

Figure 4.

This figure has been redrawn with larger numbers to make easier to read. In addition, panel (c) has been changed from showing a dropsonde from Flight B737 to a second dropsonde from Flight B734 in response to a minor correction from the external examiner. This is because flight B737 is not referred to anywhere else in the manuscript, and so its inclusion here is confusing. All comparisons between model and dropsonde (including for flight B737) are included in the supplementary material, and the figures have been redrawn to be clearer.

Figure 5.

Titles for the different panels have been added, and the caption has been edited to make it clearer what each panel is, in response to Reviewer #2, comment #8 and Reviewer J. Reid, comment #5.

Figure 6.

This figure has been redrawn with the standard model run and modified emissions run on the same panels, as recommended by Reviewer #2, comment #9.

Figure 9.

This figure has been redrawn with updated data from the flight campaign for the values of single scattering albedo. This is due to an error in the calculation of single scattering albedo in the discussions paper. This results in slightly lower single scattering albedo measured by the flights. The conclusions drawn from this figure do not change.

Figure 10.

Panels showing the volume distribution have been added, in response to Reviewer J. Reid, comment #6.

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Characterising Brazilian biomass burning emissions using WRF-Chem with MOSAIC sectional aerosol

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Abstract. The South American Biomass Burning Analysis (SAMBBA) field campaign took detailed in-situ flight measurements of aerosol during the 2012 dry season to characterise biomass burning aerosol and improve understanding of its impacts on weather and climate. Developments have been made to the Weather ~~research~~ Research and Forecast model with chemistry (WRF-Chem) model to improve the representation of biomass burning aerosol in the region by coupling a sectional aerosol scheme to the plume rise parameterisation. Brazilian Biomass Burning Emissions Model (3BEM) fire emissions are used, prepared using PREP-CHEM-SRC, and mapped to CBM-Z and MOSAIC species. Model results have been evaluated against remote sensing products, AERONET sites, and four case studies of flight measurements from the SAMBBA campaign.

WRF-Chem predicted layers of elevated aerosol loadings (5–20 $\mu g\,sm^{-3}$) of particulate organic matter at high altitude (6–8 km) over tropical forest regions, while flight measurements showed a sharp decrease above 2–4 km altitude. This difference was attributed to the plume-rise parameterisation overestimating injection height. The 3BEM emissions product was modified using estimates of active fire size and burned area for the 2012 fire season, which reduced the fire size. The enhancement factor for fire emissions was increased from 1.3 to 5 to retain reasonable aerosol optical depths (AOD). The smaller fire size lowered the injection height of the emissions, but WRF-Chem still showed elevated aerosol loadings between 4–5 km altitude. Over eastern Cerrado (savannah-

like) regions, both modelled and measured aerosol loadings decreased above approximately 4 km altitude.

Compared with MODIS satellite data and AERONET sites, WRF-Chem represented AOD magnitude well (between 0.3–1.5) over western tropical forest fire regions in the first half of the campaign, but tended to over-predict them in the second half, when precipitation was more significant. Over eastern Cerrado regions, WRF-Chem tended to under-predict AOD. ~~Modeled~~ Modelled aerosol loadings in the east were higher in the modified emission scenario. The primary organic matter to black carbon ratio was typically between 8–10 in WRF-Chem. This was lower than western flights measurements (interquartile range of 11.6–15.7 in B734, 14.7–24.0 in B739), but similar to the eastern flight B742 (8.1–10.4). However, single scattering albedo was close to measured over the western flights (0.87–0.89 in model; ~~0.88~~0.86–0.91 in flight B734, and ~~0.86~~0.81–0.95 in flight B739 measurements) but too high over the eastern flight B742 (0.86–0.87 in model, ~~0.81~~0.79–~~0.84~~ .82 in measurements). This suggests that improvements are needed to both ~~modeled~~ modelled aerosol composition and optical properties calculations in WRF-Chem.

1 Introduction

Biomass burning in South America is a globally significant source of carbonaceous aerosol (black carbon (BC) and organic carbon (OC)) (Streets et al., 2004). As well as seriously impacting on the health of the local population (Ignotti et al., 2010; de Andrade Filho et al., 2013), this biomass burning aerosol (BBA) influences the climate on a regional and global scale (Andreae et al., 2004; Zhang et al., 2009; Boucher et al., 2013). BBA can impact weather and climate directly, through interaction with radiation (Haywood and Boucher, 2000), and indirectly, by acting as cloud condensation nuclei (CCN), changing cloud optical properties, lifetime and capacity to initiate precipitation (McFiggans et al., 2006). Aerosol optical properties and suitability as CCN are both highly sensitive to the size distribution and composition of the aerosol population (Bond and Bergstrom, 2006; Abdul-Razzak and Ghan, 2002; McFiggans et al., 2006). Modelling the impacts of BBA on a regional scale requires a fully coupled "online" approach, with detailed descriptions of the aerosol properties and two-way interactions between the aerosol, radiation and cloud processes (Wang et al., 2006; Wang and Christopher, 2006; Grell and Baklanov, 2011).

High-quality emissions are essential for running chemical transport or coupled models. PREP-CHEM-SRC is a pre-processor, designed to combine data from multiple global emission databases to produce anthropogenic, biogenic and biomass burning gridded emission maps (Freitas et al., 2011). Originally developed for the CCATT-BRAMS model (Freitas et al., 2009; Longo et al., 2010), it has been extended for use with the Weather Research and Forecast model with Chemistry (WRF-Chem, Grell et al., 2011). PREP-CHEM-SRC can generate fire emissions using either the GFEDv2 inventory to produce 8 day averages (Van der Werf et al., 2006), or daily maps using the Brazilian

Biomass Burning Emission Model (3BEM) (Longo et al., 2010). 3BEM has been shown to improve modelled predictions of CO compared to the lower-resolution GFEDv2 dataset (Longo et al., 2010).

Both of these inventories use a traditional "bottom-up" approach, whereby emissions for each species ([i]) are estimated by multiplying emission factors ($EF^{[i]}$) with an estimate of the burned biomass. Satellite data is used to quantify global fire activity in terms of fire count, observed burnt area or fire radiative power (FRP), and subsequently apply properties such as fuel load and combustion completeness from model calculations or limited field and laboratory measurements. The fire properties can be very difficult to measure, resulting in large uncertainties in the emissions (Van der Werf et al., 2010; Ichoku et al., 2012; Kaiser et al., 2012; Zhang et al., 2014). Newer, "top-down" approaches to producing fire emissions systematically include information from large-scale smoke plume observations, e.g. in flux inversion from satellite observations (Huneeus et al., 2012; Ichoku and Ellison, 2013), or enhanced aerosols in Kaiser et al. (2012). These methods show a lot of promise for being able to produce near real-time fire emissions for air quality forecasting, although there are difficulties related to the retrieval algorithms and consistency between different data sources (Pereira et al., 2009). Measurements of FRP are also generally limited to cloud-free regions, and affected by the time of satellite passover and obstructions of line of sight to the fire, for example by tall trees (Kaiser et al., 2012). This can lead to biases in fire emissions in some regions of the globe (Andela et al., 2013).

The high temperatures of open vegetation fires produce flaming emissions with ~~a lot of~~ substantial associated buoyancy. In large fires, the ~~emitted~~ rising air-mass ~~may rise far above the planetary boundary layer, in some cases inducing~~ can induce convection forming so-called pyrocumulus clouds which inject emissions high above the planetary boundary layer (Andreae et al., 2001). The height of the plume can vary hugely, depending on season, the biome being burned, atmospheric stability conditions and size of fire (Val Martin et al., 2010; Sofiev et al., 2013). Many global models mix emissions within the boundary layer or specify an injection height. For example, Dentener et al. (2006) provides recommended mixing heights for different biomass burning regions for global models: ~~agricultural waste only in the lowest model levels,~~ tropical fires in the lower 1 km, temperate fires in the lower 2 km and boreal up to 6 km. ~~However~~Wang et al. (2006); Yang et al. (2013) and Wang et al. (2013) specify injection heights of 1.2, ~~larger fires may~~ 0.8 and 0.7 km for fires in Central America, Sub-Sahara and southeast Asia respectively, producing results which compare well with ground-based and remote observations. However, failing to account for the largest fires which penetrate above the boundary layer ~~. Failing to account for these~~ may result in the underestimation of emissions ~~in~~ into the free troposphere (Colarco et al., 2004; Ichoku et al., 2012).

A plume-rise parameterisation that can be embedded into regional transport models was developed by Freitas et al. (2007). The 1-D plume-rise parameterisation was initially implemented in the CCATT-BRAMS model (Freitas et al., 2009; Longo et al., 2010). Freitas et al. (2007) have shown improved representation of the vertical profile of carbon monoxide (CO) compared to measurements

from the 2002 SMOCC campaign when using the plume-rise parameterisation. This parameterisation has been successfully ported into WRF-Chem (Grell et al., 2005), to be used with the RADM (Stockwell et al., 1990) or RACM (Stockwell et al., 1997) chemical mechanisms, and GOCART (Chin et al., 2000) or MADE/SORGAM (Ackermann et al., 1998) aerosol. It has been used in many studies, for example to investigate the impact of Alaskan wildfires on weather forecasts (Grell et al., 2011); to study the effects of BBA on clouds, deep convection and precipitation in the Amazon (Wu et al., 2011a, b); and evaluating the impact of fire emissions on ozone (O_3) formation (Bela et al., 2015).

While improvements have been observed when using the plume-rise parameterisation in some studies, care should be taken. There are difficulties in using a parameterisation to represent such a complex non-linear process, as the properties needed (such as fire size, buoyancy and entrainment rate) are difficult to ~~quantify,~~ constrain, and in some cases impossible to measure, potentially leading to large errors (Ichoku et al., 2012). Indications of the plume-rise over-predicting injection height have been observed. For example, Wu et al. (2011a) found clear-sky aerosol extinction levels between 800 and 100 hPa to be higher in WRF-Chem when comparing against CALIPSO satellite observations, although they were unsure how much of this discrepancy was due to the plume-rise parameterisation and how much from convective transport. ~~found over 95of North American tropical forest fires plume injection heights measured using the MISR satellite to be less than 1.5~~Fig. 5 in Sofiev et al. (2013) shows most Amazonian plumes to be below 2.5 km, while Fig. 3 in Freitas et al. (2011) ~~shows modelled~~ models mid-afternoon ~~South-America tropical forest emissions to have~~ South-American tropical forest injection heights between 4 and 9 km.

Having aerosol injected into the wrong portion of the vertical column can have many implications. Accurate injection height is required to capture long-range transport of fire emissions (Colarco et al., 2004). The main loss-processes for BBA are wash-out and wet-deposition (Taylor et al., 2014), therefore aerosol above cloud will likely remain in the atmosphere for longer and be transported further from source. In addition, the effect of BC on atmospheric heating rates is different at different altitudes, becoming more important aloft (Samset and Myhre, 2011; Ban-Weiss et al., 2011; Samset et al., 2013).

This study aims to critically evaluate the plume-rise parameterisation in WRF-Chem against in-situ flight measurements over Brazil. The work has been carried out as part of the South American Biomass Burning Analysis (SAMBBA) project, an international collaboration set up to better understand and reduce the uncertainties associated with the impacts of biomass burning in South America on regional and global climate, air quality, and ecosystems. The observational phase of SAMBBA consisted of an airborne measurement campaign using the UK Facility for Airborne Atmospheric Measurement (FAAM) BAe-146 research aircraft (Morgan et al., 2013), alongside a longer term ground based deployment (Brito et al., 2014).

The SAMBBA modelling campaign consists of a hierarchy of models across a range of scales, from the cloud-resolving to the global. WRF-Chem is being applied to better understand the properties and impacts of BBA at a regional scale. This study describes developments being made to the WRF-Chem model to improve the applicability of the model for this task. The MOdel for Simulating Aerosol Interactions with Chemistry (MOSAIC) (Zaveri et al., 2008) aerosol mechanism has been used with the plume-rise parameterisation in order to improve the physical description and size distribution of modelled BBA. Work has also been conducted to modify the input parameters used by the 3BEM emissions and the plume-rise parameterisation in order to better control the injection height of BB emissions.

Model runs in this study have been carried out using a modified version of WRF-Chem v3.4.1. Model results are critically assessed against remote measurements of aerosol optical depth (AOD), from satellites and ground based AERONET stations (Holben et al., 2001), and in-situ measurements from the BAe-146 aircraft campaign. This is aimed at characterising the horizontal and vertical distribution of the regional haze, evaluating the behaviour of the plume-rise parameterisation, and comparing the composition, size distribution and optical properties of the aerosol population with a high-resolution data source. With the aerosol distribution and properties characterised, the model ~~setup~~ configuration can be justifiably used to investigate the impacts of the aerosol on regional weather and climate in future studies.

2 Model, emissions and the plume-rise parameterisation description

2.1 WRF-Chem and the sectional MOSAIC aerosol mechanism.

WRF-Chem is a regional, fully-coupled "online" model (Grell et al., 2005), where all prognostic meteorological, chemical and aerosol variables are integrated on the same timestep and are transported using the same advection and physical parameterisations. This makes it ideal for investigating the impacts of atmospheric composition on weather at a regional scale (Grell and Baklanov, 2011; Baklanov et al., 2014). ~~There are several aerosol mechanisms available in WRF-chem. Of these, only MOSAIC uses the more rigorous~~ For this study the MOSAIC aerosol (Fast et al., 2006; Zaveri et al., 2008) and CBM-Z gas-phase (Zaveri and Peters, 1999) mechanisms are used. MOSAIC uses a sectional representation of aerosol size distribution, ~~enabling~~ with detailed aerosol interactions with radiation and clouds described by (Chapman et al., 2009). ~~MOSAIC is only compatible with a subset of chemical mechanisms in WRF-Chem. For this study, the gas-phase mechanism used is CBM-Z~~ The Modal Aerosol Dynamics model for Europe (MADE) scheme (Ackermann et al., 1998) has also been used with WRF-Chem for investigating aerosol–radiation–cloud interactions (e.g. Grell et al., 2011; Wu et al., 2011b). However the sectional MOSAIC scheme allows for a more nuanced representation of particle composition variation across 8 size bins as opposed to 3 modes, and does

not a priori assume log-normal aerosol size distributions. It is, however, significantly more expensive to run than the modal scheme.

The aerosol size distribution in MOSAIC is described by 8 size bins spanning a dry particle diameter (D_p) range of 39 nm to 10 µm ~~. In 8-bin mode the bin bounds increase geometrically by a factor of two for each bin, as shown in~~ (see Table 1). The chemical constituents of the aerosol are assumed to be internally mixed within ~~bins~~each bin, and externally mixed between bins. MOSAIC carries five inorganic ions, plus three other aerosol species: black carbon (BC); particulate organic mass (POM); and other inorganics (OIN), which includes crustal and dust particles (Zaveri et al., 2008). Secondary organic aerosol (SOA) has been incorporated into MOSAIC using the volatility basis set (VBS) (Shrivastava et al., 2011, 2013). However, this ~~is still under development and aerosol-radiative interactions have not yet been included. It is currently unclear how much of an impact including SOA formation has on aerosol composition in regions heavily effected by biomass burning emissions. Some recent evidence suggests that, after the first few minutes of ageing, there is little SOA formation . For example, show the POM:CO ratio is conserved downwind of fires and likely determined at source, depending on the fuel type and burning conditions. However, , found SOA formation in the first 2–4h of plume ageing to be a significant contributor to BBA composition and properties.~~ was thought to be experimental at the time of study and so not used. Further work is ongoing to incorporate the VBS to study SOA formation and impacts over the SAMBBA period.

~~The optical properties of an aerosol population depend on the chemical composition and size distribution . Interactions with radiation are most efficiently when the diameter is of the same order as the wavelength (λ) of the incident light, typically a few hundredfor visible light . The most~~ The most important chemical component in determining ~~optical properties~~ aerosol radiative absorption is BC, due to the high imaginary component of its complex refractive index ($1.95 - 0.79i$ at 550 nm, as recommended by Bond and Bergstrom, 2006). The absorbing properties of BC can be enhanced by the non-absorbing aerosol components with which it is mixed (Bond et al., 2006, 2013). To simulate this, a "mixing-rule" is employed to calculate the bulk complex refractive index of each bin ~~. Three mixing-rules are available in WRF-Chem: volume averaging, where the BC is evenly mixed with the other components; Maxwell–Garnet, where the BC is seen as small, randomly distributed particles; and Shell–Core, where the BC forms the core of each particle, surrounded by a "shell" of everything else.~~ (Ackermann and Toon, 1982). Bond et al. (2006) strongly recommend not using a volume-averaging mixing rule, as it tends to artificially overestimate the absorption enhancement of BC. For this study a Maxwell-Garnet mixing-rule has been used. This treats the BC as small particles randomly distributed within a well mixed matrix composed of the other chemical components.

Mie calculations are used to first find the optical properties of each bin (Toon and Ackerman, 1981), then summed over all bins to give the bulk optical properties of the aerosol population: the extinction, (b_{ext}), scattering coefficient (b_{scat}), absorption coefficient (b_{abs}), single scattering albedo (ω_0) and asymmetry factor for scattering (g). Each of these is defined as a function of λ, with ω_0

being the ratio of scattering to extinction:

$$\omega_0 = \frac{b_{\text{scat}}}{b_{\text{scat}} + b_{\text{abs}}} = \frac{b_{\text{scat}}}{b_{\text{ext}}}. \quad (1)$$

Full descriptions of the aerosol optical calculations in WRF-Chem are described by Fast et al. (2006) and Barnard et al. (2010).

~~Recent WRF-Chem developments have enabled explicit modelling of the mixing state of BC with other components . These have shown internal mixing-rule approximations to overestimate the radiative absorption of aerosol by 30–40in the boundary layer. This treatment is, however, extremely computationally expensive to run.~~

~~Within WRF-Chem it is assumed that the organic fraction of the aerosol is non-absorbing in the short-wave. However, there is some evidence of POM, particularly in BBA, weakly absorbing radiation at some wavelengths . suggest BrC absorption can be parameterised using a relation between the POM:BC ratio. Weak short-wave absorption by POM may need to be added in future versions of WRF-Chem to model BBA optical properties accurately.~~

2.2 Brazilian biomass burning emissions model

The 3BEM fire emissions product uses daily data of detected fires from several satellite products: the Moderate Resolution Imaging Spectroradiometer (MODIS) (Giglio et al., 2003), the Geostationary Operational Environmental Satellite-Wildfire Automated Biomass Burning Algorithm (GOES WFABBA, cimss.ssec.wisc.edu/goes/burn/wfabba.html; Prins et al., 1998) and the Brazilian National Institute for Space Research (INPE) fire product, which uses the Advanced Very High Resolution Radiometer (AVHRR) onboard the NOAA polar orbiting satellite series (www.cptec.inpe.br/queimadas; Setzer and Pereira, 1991). A filter algorithm that removes fires within 1 km of each other is used to prevent double counting between datasets (Longo et al., 2010).

Each fire pixel is cross-referenced against 1 km resolution maps of vegetation and land-use for the year 2000 (Olson et al., 2000; Sestini et al., 2003). The fire is assigned one of four ~~different~~ biomes: tropical forest, extra-tropical forest, savannah/~~cerrago~~Cerrado, or grassland. ~~Different~~ Each biome has an associated carbon density (~~α~~α_{veg}) and combustion ~~factors (β) are used~~ factor (β_{veg}). Emission factors ($\text{EF}^{[i]}_{\text{veg}}$) for each biome type ~~, and are multiplied to find the total burned biomass. Each biome type also has associated emission factors ($\text{EF}^{[i]}_{\text{veg}}$) of , to convert from mass of biomass burned to quantity of each emitted species (i).~~ are taken from Andreae and Merlet (2001). These are further scaled by an estimated total burned area (A_{fire})~~. The burned area~~ , which cannot be directly measured from satellite products in real time, although it may be estimated from fire radiative product (FRP) ~~using an algorithm,~~ if suitable data is available. ~~An average area burned is often used due to difficulties in quickly retrieving accurate readings from satellite products.~~ Some fires detected by the WFABBA product have A_{fire} estimated using the Dozier method (Dozier, 1981, http://wfabba.ssec.wisc.edu/ongoing.html). If this ~~data~~ is not available (as is the case for ~~fire~~ fires

detected with the MODIS and INPE products), ~~an average fire size~~ a burned area of 22.8 ha is used for all vegetation types (Longo et al., 2010). Finally, the fire emissions may need to be scaled up by an enhancement factor (f_x) in order to account for uncertainties and produce physically realistic aerosol optical depths (AODs). These factors are combined to give the emitted mass ($M^{[i]}$) of each species [i]:

$$M^{[\mathrm{i}]} = \alpha_{\mathrm{veg}} \cdot \beta_{\mathrm{veg}} \cdot \mathrm{EF}^{[\mathrm{i}]}_{\mathrm{veg}} \cdot A_{\mathrm{fire}} \cdot f_x. \quad (2)$$

By default, f_x is set to 1.3 for South American fires in PREP-CHEM-SRC v1.4. Enhancement factors such as this have been applied to many emission products and models, in order to bring bottom-up inventories in line with top-down constraints (Kaiser et al., 2012; Tosca et al., 2013)~~fire emission inventories. Values~~ . Values of f_x in the literature typically range from 2 to 5. For example, Wu et al. (2011a) multiplied 3BEM OC and BC surface aerosol emissions by a factor of 5 when simulating the 2006 fire season~~.~~ , Tosca et al. (2013) used an enhancement factor of 2.4 for South American fires using the GFEDv3 inventory with the CAM-5 model, and Kaiser et al. (2012) recommend scaling GFASv1.0 particulate emissions by a factor of 3.4. The need for this factor highlights the difficulties and uncertainties in estimating fire emissions using ~~the currently available~~ current observations and understanding~~of the processes involved. demonstrate that the required factor depends strongly on the underlying emission inventory and the geographical location.~~ . Zhang et al. (2014) have shown existing emission inventories can differ by a factor of 10 in some locations, although top-down estimates tend to show less variation.

2.3 Plume rise parameterisation

The Freitas et al. (2007) plume-rise parameterisation applies a 1-D ~~cloud-resolving~~ cloud-parcel model to each grid-column within the WRF-Chem model domain that contains a fire. ~~It calculates the~~ The full set of equations are described in detail by Freitas et al. (2007, 2010). The parameterisation calculates an initial plume buoyancy ~~by estimating the energy and moisture released from the fire, based on fire size and carbon density, and using~~ which depends on biome burned (with forest fires releasing more heat than savannah or grassland fires) and ambient environmental conditions along the column retrieved from the parent model. ~~The plume rises until it becomes dynamically stable, and the height at this point is passed to the parent model. The~~ The microphysical parameterisation of Kessler (1969), with accretion and ice formation of Ogura and Takahashi (1971), is used to compute whether convection occurs ~~,~~ and the latent energy released ~~and the effect on the height of the plume, using an initial cloud condensation nuclei (CCN) concentration of 10^5taken from . The total fire emissions are split between smouldering and flaming phases, with the fraction apportioned depending on the vegetation being burned. A lower and upper estimate~~ if so. Lower and upper estimates of heat flux ~~is~~ are used to give lower and upper limits of the injection height. The ~~flaming fraction is emitted between these~~ total fire emissions are split between smouldering and flaming

phases, with the flaming fraction emitted between the elevated injection heights, while smouldering emissions are emitted into the lowest mode level. .

The behaviour of the plume-rise parameterisation is dependent on the location and size of each fire. The heat flux and flaming fractions of the fires differ for each of the four biomes, with forest fires burning more energetically than savannah or grassland fires due to higher carbon density . The The main loss of buoyancy results from entrainment of colder air from the surrounding environment the surrounding air into the plume:

$$\frac{\partial w}{\partial t} + w\frac{\partial w}{\partial z} = -(\lambda_{\text{entr}} + \delta_{\text{entr}})w, \tag{3}$$

where w is the vertical speed of the plume, and λ_{entr} and δ_{entr} are the lateral and shear entrainment terms respectively. λ_{entr} is given by:

$$\lambda_{\text{entr}} = \frac{2\alpha}{R}|w|, \tag{4}$$

where R is the radius of the plume, w the vertical velocity of the plume and α the dynamic entrainment constant (Freitas et al., 2007), taken to be 0.05 for good agreement with the Active Tracer High resolution Atmospheric Model (ATHAM) model simulations (Freitas et al., 2010). Freitas et al. (2010) have expanded the parameterisation to include entrainment of shear wind as well as vertical:

$$\delta_{\text{entr}} = \frac{2}{\pi R}(u_{\text{e}} - u), \tag{5}$$

where u and u_{e} are the horizontal wind speeds of the plume and environmental respectively. Note that $(u_{\text{e}} - u)$ in Eq. (5) is formulated as a scaler difference. This implicitly assumes , implicitly assuming the environmental and plume winds are in the same direction. A vector difference would account for changes in wind direction in the vertical column. The current formulation therefore systematically underestimates the horizontal entrainment effect, although the difference is likely to be small in most cases.

The plume radius R is derived from the active size of the fire (S_{fire}), assuming the cross-section of the plume to be circular (i.e. $R \propto \sqrt{S_{\text{fire}}}$). As both λ_{entr} and δ_{entr} are inversely proportional to R, larger fires undergo less entrainment and have higher injection heights (Freitas et al., 2010). The full set of equations for plume dynamics, microphysics and entrainment are described in detail by .

3 Model and emission product developments

This section of the paper presents development work carried out to improve BBA representation within WRF-Chem with sectional aerosol. The developments can be summarised as: are: modification of PREP-CHEM-SRC was modified to use updated fire size data for the 2012 fire season when generating 3BEM emissions . This information was fed to the plume-rise parameterisation to achieve more realistic injection heights.

~~Gas-phase emissions from PREP-CHEM-SRC were mapped to~~ to update fire size and area; mapping PREP-CHEM-SRC emissions to CBM-Z ~~species, and aerosol emissions to MOSAICsectional aerosol with appropriate size distributions.~~

~~Boundary conditions derived~~ and MOSAIC; and deriving boundary conditions from the MACC-II product ~~were added to capture long-range~~ in order to capture long range transport of BBA~~into the regional model domain.~~ .

3.1 Updating fire size estimates for the 2012 biomass burning season

The plume-rise parameterisation in WRF-Chem shows a tendency towards overestimating the injection height of flaming emissions, as will be shown in the results section in this paper. Ichoku et al. (2012) suggest restraining the plume height using remote measurements of plume height, such as the MISR satellite. For this work, the inputs of the parameterisation have been refined ~~in~~ with the aim of improving the predictive capacity of the injection height calculation.

There are several assumptions built into the 3BEM emissions and plume-rise setup which may make it prone to having a positive bias. Firstly, there has been a downward trend in fire emissions since the late 1990s and early 2000s (Artaxo et al., 2013). Much of the evaluation of the plume-rise parameterisation and 3BEM emissions product has used data from 2002 (Freitas et al., 2007, 2009; Longo et al., 2010). In using the relatively large ~~estimate of the~~ average burned area ~~for all fires of~~ estimate of 22.8 ha, we may be simulating ~~overly~~ large fires more representative of the previous decade than the modern day. Secondly, the active fire size (S_{fire}) used by the plume-rise parameterisation is ~~the same as the~~ equal to the total burned area (A_{fire}) used to calculate the emitted mass~~(i. e. $A_{\text{fire}} = S_{\text{fire}}$). Logically, it~~ . It is not reasonable to assume that the actively burning portion of a fire is the same as the total burned area. ~~It is known that fires~~ Fires are known to spread along a front (Viegas, 1998), and this behaviour should be approximated in the equations used to calculate the plume-rise.

A number of methods for deriving fire size from satellite products have been developed. Dozier (1981) proposed a bi-spectral approach that utilises the estimated radiance at 4 and 11 μm. However, inaccuracies in data acquisition and the digital processing required (for example, co-registration between bands with distinct spatial resolutions and point spread functions, sensor noise and spectral atmospheric interference) could generate large errors in fire size estimation (Giglio and Kendall, 2001; Giglio and Justice, 2003). As a consequence a number of modifications to the Dozier method have been proposed ~~.~~ (Peterson and Wang, 2013; Peterson et al., 2013; Shimabukuro et al., 2013; Giglio and Schroeder, 2014). Peterson et al. (2014) have developed a probabilistic method for estimating the emission injection height based on FRP and retrieved burned area products from MODIS for use over boreal forests. However, fires which occur within the biomes specific to the Amazon and Cerrado regions present distinct behaviours (Arai et al., 2011) for which the majority of these schemes have not been calibrated and validated.

For this study, updated estimates of burned area for the 2012 season have been used, acquired from a pre-operational product of CPTEC/INPE (Shimabukuro et al., 2013). In this product, burned area and active fire size are estimated through FRP and Fire Radiative Energy (FRE) based coefficients to different types of vegetation in South America (grassland, herbaceous, scrublands, forest, and agriculture), derived from simultaneous observations of Thematic Mapper (TM) and Enhanced Thematic Mapper Plus (ETM+) images of Landsat 5 and Landsat 7, respectively. MODIS FRP values were used to estimate the fire size using:

$$\mathrm{GRID}_{(\mathrm{lon,lat,FRP,LULC})} = \sum_{x=-\alpha}^{\alpha} \sum_{y=-\beta}^{\beta} (\vartheta(x,y)\mathrm{FRP}(\mathrm{lon}+x,\mathrm{lat}+y) \cap \vartheta(x,y)\mathrm{LULC}(\mathrm{lon}+x,\mathrm{lat}+y))A_{\mathrm{c}}, \quad (6)$$

where $\vartheta(x,y)$ represents the convolution mask of $M \times N$ size (rows $\times$ columns), FRP is the estimated MODIS FRP derived from MOD14 and MYD14 products, LULC is the land cover type derived from MCD12Q1 product, and A_{c} is the fire size coefficient (0.00021–0.00029 $\mathrm{km^2\,MW^{-1}}$). GRID is the fire size (S_{fire}) defined for all points in which the mask of $M \times N$ size completely overlaps the grid ($\mathrm{lon} \in [\alpha, M-\alpha]$, $\mathrm{lat} \in [\beta, N-\beta]$). The same approach is applied to derive A_{fire} by replacing FRP with FRE, as described in Shimabukuro et al. (2013).

Table 2 shows estimates of mean A_{fire} and S_{fire} for the 2012 Brazilian fire season, made using the above method. The estimates are dependent on biome (in a similar fashion to $\mathrm{EF}^{[\mathrm{i}]}_{\mathrm{veg}}$, α_{veg} and β_{veg} in Eq. 2). As the data was collated for South America over 2012, it should provide more representative estimates of burned area and fire size for the SAMBBA study, given the downward trend in fires over the past decade. S_{fire} is some 10 to 20 times smaller than 22.8 ha, depending on the biome, meaning the entrainment rate is increased by a factor between 3 and 5. The modified 3bem_emissions.f90 code for PREP-CHEM-SRC v1.4 is included in the Supplement, with instructions on how to modify for another campaign.

Reducing the estimated A_{fire} to a more reasonable size also reduces the total emitted mass. It was found that this resulted in unrealistically low aerosol optical depths (AODs). Previous models have used higher factors to get reasonable AODs as discussed ~~above~~in Section 2.2. For this study f_x has been increased from 1.3 to 5. This has been estimated based on the reduction of tropical forest A_{fire} by approximately a factor of 5 from the original default area of 22.8 ha, while the other biomes are between a third and half the size. As forest fires are the dominant source of emissions in the region, this maintains similar magnitudes of particulate emissions so the study can focus on the implications of the injection height changes.

3.2 Coupling PREP-CHEM-SRC emissions with CBM-Z MOSAIC

The emissions generated by PREP-CHEM-SRC are made with the RADM2 and GOCART speciation. For the gas-phase emissions we have ported the mappings used for anthropogenic RADM2

speciations to CBM-Z within WRF-Chem. The excess carbon from longer chained hydrocarbons are added to the CBM-Z species PAR, OLET and OLEI, as described in Zaveri and Peters (1999). ~~BBA emissions for MOSAIC have been treated differently from anthropogenic emissions within the emission subroutine. While anthropogenic emissions are only injected into the lower most levels, BB emissions loop through the entire vertical column in order to distribute flaming emissions~~ Biomass burning flaming emissions are distributed within the model vertical column using the injection heights calculated by the plume-rise parameterisation.

Emissions of BBA are usually observed in two size modes, a sub-micron accumulation mode which makes up the majority of the particulate number and mass, plus a coarse mode made up of a lower number of larger particles (Reid and Hobbs, 1998). The fine mode is mostly organic compounds, with around 10 % BC and inorganic species respectively. The coarse mode is made up of dust, ash, carbon aggregates and unburned fuel (Reid et al., 2005; Janhäll et al., 2010). PREP-CHEM-SRC produces emission values for BC, OC, $\mathrm{PM}_{2.5}$ and PM_{10}, based on the factors in Andreae and Merlet (2001). For this study all BC and OC are assumed to be included in the $\mathrm{PM}_{2.5}$ fraction of emissions. ~~The emissions of Organic Carbon (OC) need to be converted to total particulate organic matter (POM), which includes the associated oxygen, hydrogen and other elements.~~ Biomass burning OC emissions have been converted to ~~POM~~Particulate Organic Matter (POM), multiplying by a factor of 1.5, following Reid et al. (2005). Similarly anthropogenic OC emissions have been multiplied by a factor of 1.6 (Turpin and Lim, 2001) to yield POM. All emitted particulate mass that is not BC or POM is assumed to be unreactive inorganic in composition, and mapped to other inorganics (OIN).

Evidence from measurements of very fresh plumes suggest that in the few seconds after burning, there are a large number of small particles which rapidly coagulate (Reid and Hobbs, 1998). After a few minutes, the distribution generally has a single large accumulation mode, sometimes with a smaller coarse mode (Janhäll et al., 2010). ~~Recent measurements of suggest significant~~ Some measurements suggest changes to CCN, size distribution and ω_0 occur over the first 2–4 ~~h~~ hours of ageing through SOA formation ~~in South African biomass burning plumes~~(Reid et al., 1998; Vakkari et al., 2014). However, these processes cannot ~~currently~~ be parameterised within this version of the model. A geometric mean diameter (D_g) of 117 nm, with a geometric standard deviation (σ_g) of 1.7, has been used to create a log-normal size distribution based on the average of 20 data points of fresh (no more than a few minutes old) smoke samples taken across several studies, compiled by Janhäll et al. (2010). This number distribution was converted to a volume distribution, normalised and, assuming a constant particle density, mapped to the 8 MOSAIC size bins. The fraction of total aerosol emissions assigned to each bin is shown in Table 1.

Biomass burning events exhibit a strong diurnal cycle (Giglio, 2007). To approximate this diurnal variation in a model, a ~~gaussian~~ Gaussian distribution with peak at a local time of around 15:00 LT (approximately 18:00 UTC over Brazil) is often used (Kaiser et al., 2009; Freitas et al., 2011). As

a large landmass such as South America spans several time zones, for this work a local time (t_l) for each emission point is calculated:

$$t_l = t_{UTC} + \frac{LON}{15} \quad (7)$$

where LON is the local longitude, in degrees, varying between $-180°$ and $+180°$. This is used to define a ~~gaussian~~ Gaussian function, $r(t_l)$, based on that used by Freitas et al. (2011), with a peak at 15:00 LT, defined such that the integral of $r(t_l)$ over 24 h is equal to 1. This function modulates the magnitude of the emissions online within WRF-Chem. While Giglio (2007) suggest different diurnal cycles in different regions of Brazil based on different biomes, it was considered problematic to extrapolate from the regions used in the study to the biomes used in PREP-CHEM-SRC, and so the single diurnal cycle of Freitas et al. (2011) was retained.

3.3 MACC-II boundary conditions

Whilst regional models benefit from the increased resolution allowed by simulating a smaller area, they are dependent on boundary conditions from global model datasets for everything occurring outside the domain bounds. There is evidence for dust and BBA from Africa being transported across the Atlantic to Brazil (Rizzo et al., 2013; Brito et al., 2014). Amazonian fire plumes may also be transported out of and recirculated back into the domain. In order to avoid simulating the whole of the Atlantic and Africa, as was done by Freitas et al. (2009), it is necessary to be confident that the emission and long-range transport of these events is well captured by ~~our~~ the boundary conditions.

The series of GEMS, MACC and MACC-II (Monitoring Atmospheric Composition and Climate – Interim Implementation; Hollingsworth et al., 2008; Flemming et al., 2013) projects have developed analysis, reanalysis and forecast products that use the MOZART-3 chemical transport model (Emmons et al., 2010) with the ECMWF Integrated Forecast System (IFS), which has been expanded to integrate measurements of reactive gases (Stein et al., 2012), greenhouse gases and aerosol (Benedetti et al., 2009) in the ECMWF 4D-Var assimilation system (see Stein et al., 2012; Inness et al., 2013, and references therein). ~~MODIS retrievals of aerosol optical depth at 550are used to constrain modelled aerosol, improving its spatial distribution . Satellite retrieval columns of reactive gases (, , , and) are also assimilated .~~It calculates aerosol and reactive gas sources, chemical conversion, transport and deposition online, i.e. at each model time step (Morcrette et al., 2009; Stein et al., 2012). Daily biomass burning emissions of the Global Fire Assimilation System (GFAS) (Kaiser et al., 2009, 2012) are also used. Using daily fire emissions and satellite assimilation gives better constraint on the chemical and aerosol loadings, providing more reliable boundary conditions.

~~Only a subset of chemical species thought to be significant in long-range transport and chemistry are included in the~~ The mapping of MACC-II ~~product: CO, , , , , , , , isoprene, peroxyacetyl nitrate (PAN) and formaldehyde (). The aerosol module used in MACC-II is described by . Five species of aerosol are carried: natural sea salt (SU) and dust (DU), and three anthropogenic aerosol (POM,~~

~~BC and SULF). SULF, POM and BC are each treated as bulk aerosol, with BC and POM treated as two components – hydrophobic and hygrophilic. SS and DU are each represented with by bins with boundaries at 0.03, 0.5, 5 and 20diameter for SS and 0.03, 0.55, 0.9 and 20for DU .~~

~~The model uses log-normal distributions with parameters of mean diameter (D_p) and geometric standard deviation (σ) as defined below (Jean-Jaques Morcrette, personal communication, 2013): SS: two log-normal distributions; the first with $D_{p,1}$=0.389, $\sigma_{p,1}$=1.9, $N_{\mathrm{tot},1}$=70, the second with $D_{p,2}$=3.984, $\sigma_{p,2}$=2.0, $N_{\mathrm{tot},2}$=3.~~

~~DU: a single log-normal distribution, $D_p = 0.58$, $\sigma_p = 2.0$.~~

~~The bulk aerosol BC, POM and is assumed to be in an accumulation mode with single log-normal distribution, D_p=0.071, σ_p=2.0. The fraction of each MACC-II bin to be partitioned into each MOSAIC bin is given by the fraction of each distribution that falls between each MOSAIC bin boundary. As the upper limit of MOSAIC aerosol is 10, all aerosol mass from the distributions above 10is discarded. See Table 3 for full apportionment to each MOSAIC size bin.~~

~~The SULF carried in MACC-II is assumed to be ammonium sulphate () when mapped to the~~ products to WRF-Chem ~~MOSAIC species , in order for the aerosol to have neutral acidity. Likewise, SS is assumed to be and is split between the and ions. The MACC-II boundary conditions were interpolated to the model grid using a modified version of the mozbc script ().~~ species is detailed in Appendix A.

4 Campaign description

The SAMBBA aircraft campaign was based in Porto Velho, northern Rondônia. This is a region with extensive biomass burning owing to forest clearance. The ground measurement site was also located in the city, upwind of urban emissions. Nineteen flights were conducted between the 14 September and 3 October 2012, encompassing not only an extensive geographic area, but also differing synoptic conditions (see Darbyshire et al., ~~2014~~in prep., for further details). Flights over the western regions encompassed two meteorological regimes as discussed in Brito et al. (2014), with Phase I (6 to 22 September 2012) representative of dry season conditions and Phase II (after 22 September) of the transition to the wet season. ~~Comparatively, conditions remained~~ Conditions remained comparatively dry throughout in the eastern Cerrado region.

4.1 Observational datasets

In this study, WRF-Chem model results are compared against various remote sensing and ground based datasets. The Tropical Rainfall Measuring Missions (TRMM) is a NASA project aiming to provide satellite derived estimates of tropical precipitation across the globe. The 3B42 product produces 3 hourly merged high quality, infrared and microwave precipitation estimates at $0.25° \times 0.25°$ resolution between $50°$ N and $50°$ S (Huffman et al., 2001, 2013).

The Moderate Resolution Imaging Spectrometer (MODIS) instrument, on board the two NASA satellites Aqua and Terra, ~~provide~~ provides measurements of Aerosol Optical Depth (AOD) across a wide spectral range at ~~$1.0° \times 1.5°$~~ $1.0° \times 1.0°$ (Remer et al., 2005). For this study, retrievals of AOD at 550 nm are used for verifying the model aerosol horizontal distribution. Overpasses over the study period and region of the globe were at approximately 03:00 and 15:00 UTC for the Terra satellite, and 06:00 and 18:00 UTC for the Aqua satellite. Model data was extracted at the these times when comparing against MODIS data. Over land, the MODIS AOD retrievals have an error of approximately 0.05 (Remer et al., 2005).

The Aerosol RObotic NETwork (AERONET) program is a ground-based deployment of around 100 sites, providing continuous observations of AOD at various wavelengths using the Version 2 Direct Sun Algorithm (Holben et al., 1998, 2001). AOD at 550 nm is estimated using measurements of AOD at 675 and 440 nm and the ~~Angström component~~Ångström exponant. The data has been screened for clouds; only level 2.0 quality assured data is used for this study. Under cloud free conditions, the error in measured AOD is approximately 0.01 (Holben et al., 2001). Data was retrieved for four sites over the central Brazilian region: Cuiabá (15° S, 56° W), Ji Paraná (10° S, 61° W), Porto Vehlo (8° S, 63° W) and Rio Branco (9° S, 67° W).

4.2 Instrument details

The suite of aerosol instrumentation used on the FAAM BAe-146 for this study is summarised in Table 4. The submicron nonrefractory aerosol composition was measured by an Aerodyne Research (Billerica, MA, USA) compact Time of Flight Aerosol Mass Spectrometer (cToF-AMS), as described by Drewnick et al. (2005); Canagaratna et al. (2007), and for FAAM operation by Morgan et al. (2009). For speciated mass loadings, detection limits are approximately 40 $\mathrm{ng\,m^{-3}}$ for organics (Drewnick et al., 2009), whilst combined measurement uncertainties are approximately 30 % (Bahreini et al., 2009; Middlebrook et al., 2012).

The Single Particle Soot Photometer (SP2), developed by Droplet Measurement Technologies (Boulder, CO, USA), was used to measure number and mass concentrations of refractory Black Carbon (rBC). Its operating principles are described in Stephens et al. (2003) and Baumgardner et al. (2004), with its utilisation onboard FAAM summarised by McMeeking et al. (2010). For reported mass loadings the measurement uncertainty is approximately 30 % (Schwarz et al., 2008; Shiraiwa et al., 2008).

Aerosol total scattering coefficients were measured by a TSI Inc (St. Paul, MN, USA) 3-wavelength integrating nephelometer (Anderson et al., 1996), with standard corrections applied for angular truncation and non-lambertian light source errors (Anderson and Ogren, 1998; Müller et al., 2011), and for relative humidity, using the humidification factors defined for Porto Velho haze in Kotchenruther and Hobbs (1998). A Radiance Research Particle Soot Absorption Photometer (PSAP) measured the

aerosol absorption coefficient at 567 nm and standard corrections for spot size, flow rate and scattering particles were applied following Bond et al. (1999); Ogren et al. (2010) and Turnbull (2010).

Aerosol number-size distributions were measured across the 20 nm to 20 µm range by a Scanning Mobility Particle Sizer (SMPS, 20 to 350 nm; Wang et al., 1990) and a GRIMM model 1.129 Optical Particle Counter (OPC, 0.3 to 20 µm; Heim et al., 2008). Note the Grimm data used in this paper is uncorrected for the minor impact of ~~line-losses and refractive index, which is thought not to be significant for BBA below 1.0. The instrument sample is extracted through a Rosemount inlet which has been shown to measure representatively below 600for aerosol in continental polluted air masses. (J. Trembath, personal communication, 2014).~~ refractive index. A Droplet Measurement Technologies Inc. (DMT) dual column Cloud Condensation Nuclei counter (CCNc) was used to measure CCN concentrations with an approximate measurement error of 7 %. The operating principles are outlined in Roberts and Nenes (2005), whilst its utilisation onboard ~~FFAM~~ FAAM is described in Trembath (2013).

The aerosol instrumentation onboard FAAM ~~sampled~~ samples through a Rosemount inlet which, despite suffering known artefacts for larger particles, is adequate for the submicron size range ~~of aerosols~~ presented here (Trembath, 2013). All measured data have been converted into units of standard temperature and pressure. Further details on instruments, calibration protocols and quality assurance of data are provided in Darbyshire et al. (~~2014~~in prep.) and Morgan et al. (~~2014~~in prep.). Carbon monoxide (CO) was measured using an Aero-Laser AL5002 VUV resonance fluorescence gas analyser. The raw CO was calibrated in-flight.

From each instrument time series the influence of fresh plumes was removed, as to isolate the regional haze measurements, following the plume identification technique discussed in Darbyshire et al. (~~2014).~~ in prep.).

4.3 Model setup

For this study a modified version of WRF-Chem Version 3.4.1 has been used. A single lambert projection domain with 226 × 196 grid cells, at a horizontal spacing of 25 km, covers most of South America. 41 vertical levels are used, spaced to give greater resolution in the boundary layer. 1 km resolution global landuse data was provided by the United States Geological Survey (USGS), with vegetation maps updated for the Brazilian Legal Amazon Region with the PROVEG dataset updated for the year 2000 (Sestini et al., 2003; Freitas et al., 2011; Beck et al., 2013). Figure 1 shows the model domain with the USGS land use categorisations. ~~The majority of the flights for this study were conducted in RondSUPERSCRIPTonia State, between 8–12S and 60–65W, along the southern edge of the Amazon basin.~~

The chemistry ~~option used was~~ options used were the Kinetic Pre-Processor (KPP Damian et al., 2002) compiled version of CBM-Z ~~gas-phase chemistry~~ (Zaveri and Peters, 1999) with 8-bin MOSAIC aerosol and aqueous chemistry (Zaveri et al., 2008). The Maxwell–Garnett mixing-rule ap-

proximation was used to calculate optical properties of the aerosol, linked with the RRTMG longwave and shortwave radiation parameterisation (Mlawer et al., 1997; Pincus et al., 2003).

The physical parameterisations used for this study are summarised in Table 5. ~~Long-term running options, for updating sea-surface temperature~~The non-local Yonsai University (YSU) planetary boundary layer (PBL) scheme (Hong et al., 2006) defines the boundary layer height as the mixed layer height:

$$h = \mathrm{Ri}_c \frac{\theta_{va}|U(h)|^2}{g[\theta_v(h) - \theta_s} \quad (8)$$

where Ri_c is the critical bulk Richardson number ($= 0.5$), $U(h)$ is the horizontal wind speed at h, θ_v is the virtual potential temperature, θ_{va} is the virtual potential temperature at the lowest model level and θ_s is the temperature at the surface. It is solved iteratively with θ_s, as described by (Hong et al., 2006). The average mixed layer height at 17:00 LT was found to be 1873 ± 541 m over forested regions and 2912 ± 301 m over Cerrado regions; approximately 800 and ~~other fields, were activated.~~ 1 300 m higher, respectively, than the values given by Fisch et al. (2004) for forest and pasture sites in dry season Amazonia.

The operational, deterministic (high-resolution) 1 day forecasts of the European Centre for Medium-Range Weather Forecasts (ECMWF) http://www.ecmwf.int/ were used to drive the meteorology. Long-term running options, for updating sea-surface temperature and other fields, were activated. Chemical boundary conditions ~~are~~ were taken from MACC-II. ~~The meteorology, including satellite data assimilation, of the~~ The MACC-II ~~assimilation system is identical (except for its lower resolution)~~ system is an extension of ECMWF's integrated forecasting system (IFS) used for operational forecasting, which is run at a lower resolution of T255 instead of T1279. Since feedback from aerosols on the meteorology is disabled, the meteorological fields are virtually identical to the operational ~~ECMWF dataset~~meteorological forecasts, albeit with lower resolution. This ensures ~~equivalence between the meteorological and chemical boundary conditions~~consistency between the chemical and meteorological boundary conditions in this study.

PREP-CHEM-SRC v1.4 was used to generate anthropogenic and biomass burning emission maps. Anthropogenic emissions of CO, SO_2, NO_x, NH_3 and NMVOCs are derived from the Emissions Database for Global Atmosphere Research (EDGAR) version 4.0 2005 emissions at $0.1^\circ \times 0.1^\circ$ resolution (Olivier et al., 2002). Primary anthropogenic aerosol emissions of BC and OC ~~at $1^\circ \times 1^\circ$ resolution from~~ are from from the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model databases ~~are used~~ $1^\circ \times 1^\circ$ resolution (Freitas et al., 2011). Burning of residue in fields, residue and dung used as biofuels, ~~and~~ fuelwood and charcoal burning ~~were~~ was included using the ~~inventory of~~ Yevich and Logan (2003) ~~, with the application of~~ inventory, applied with Andreae and Merlet (2001) emission factors~~, and consolidated into the anthropogenic emissions input file.~~ . Modifications to PREP-CHEM-SRC were made to convert OC into POM for all anthropogenic emissions with a factor of 1.6 (based on Turpin and Lim, 2001) and include NH_3 emissions. Bio-

genic emissions were calculated "online" using the Model of emissions and Gases and Aerosols from Nature (MEGAN) version 2 (Guenther et al., 2006).

Fire emissions were calculated using the 3BEM emissions inventory. Two ~~emission~~ emissions scenarios have been used for this study:

- Standard 3BEM emissions: default $A_{\text{fire}} = 22.8\,\text{ha}$, $S_{\text{fire}} = A_{\text{fire}}$. $f_x = 1.3$.
- Modified 3BEM emissions. A_{fire} and S_{fire} depend on vegetation type, as described in Table 2. $f_x = 5$.

Figure 2 shows horizontal maps and vertical cross-sections of the plume-risen fire emissions ~~through 9S~~ for the two scenarios. ~~The four panels on the left are for Phase I, while the four right panels are for Phase II. The~~ The horizontal distribution is similar for both scenarios. There is a significant reduction in average emissions in the second phase of the campaign, along with a relative shift of emissions ~~eastwards to drier, cerrado regions. This shift in distribution is largely controlled by change in number and location of fire-pixels.~~ east towards drier, Cerrado regions. The vertical profiles of emissions show much greater differences between the two scenarios. The cerrado fires, predominantly east of 50° W, have peak injection heights of just above 4 km in both emissions scenarios~~, around the same height or just above as the daytime boundary layer~~. The western fires, which are predominantly tropical forest biomes, peak between 5 and 12 km in the standard ~~3BEM~~ emission scenario, and 3–6 km in the modified emission scenario, despite the ~~boundary layer being considerably lower~~ lower boundary layer over the forest~~(typically between 1.5 and 2). The forest emissions are higher in the model due the higher fuel load.~~ . While the injection height is ~~significantly~~ lower in the modified emissions scenario, ~~this~~ it is still higher than what is usually reported~~in the literature.~~ . For example, in a ~~review of North American tropical fire plume measurements, show 95of tropical forest~~ global review of MISR fire plume height retrievals Sofiev et al. (2013) show the majority of daytime August wildfire plumes are below ~~1.5~~2.5 km in altitude over Amazonia.

The injection height shows a strong diurnal cycle, reflecting the cycle of fire activity which follows a fixed parameterisation in this study. Flaming emissions are injected just above ground at night and the early morning/late evening. Over the course of the day, as the atmosphere becomes more unstable, the injection height for each fire will typically make a discontinuous "jump" into the higher levels of the atmosphere as and when the convection is triggered within the parameterisation. The time and height of this "jump" varies from day-to-day, depending on the ambient meteorological conditions, and is highly non-linear. ~~The behaviour of the diurnal cycle in emissions and injection height~~ This behaviour can be observed in the ~~video~~ 3D animation of model CO over the campaign ~~period~~ rendered using VAPoR (Clyne et al., 2007) included in the Supplement.

The scenarios were run from 1 September to 1 October 2012, encompassing all the flights of interest. Between 1 September 2012 and 11 September the model was ~~run~~ spun up with meteorological

nudging to build reasonable background aerosol fields in the model. From 11 September to 1 October, meteorological fields were reset from the ECMWF data every two or three days. ~~Between 1 and 14 September the model outputted data 3hourly, with the first three days ignored as spin-up. This period was needed to both give time for spin-up and because the aerosol loadings were higher at this time, providing interesting comparisons against satellite and ground-based measurements. From 14 September, model data was outputted hourly to give higher temporal resolution when comparing with flight data.~~ Nudging was turned off for the later periods so as not to interfere with aerosol radiative feedbacks (to be discussed in more detail in future studies).

5 Results and analysis

The purpose of this study is to characterise the aerosol population and compare with measurements. The aim is to develop as accurate a picture as possible of the horizontal and vertical distribution, size distribution and composition.

Prior to investigating the aerosol carried by the model, we will establish that it represents the meteorological fields with a reasonable level of accuracy. Aerosol loss processes are dominated by wet deposition, and the injection height of the flaming emissions will depend partly on the vertical profile of the atmosphere and wind speed in the column. We will then proceed into more in-depth characterisation of the aerosol, firstly over the whole period of the campaign against remote satellite measurements and long term AERONET sites, then with more detailed in-situ measurements from the SAMBBA aircraft campaign.

5.1 Verification of meteorology and stability profile of atmospheric column

Figure 3 shows maps of average precipitation over the two phases of the campaign. The two panels on the left are derived from the TRMM 3B42 product of 3 hourly gridded precipitation at $0.25^\circ \times 0.25^\circ$ resolution (Huffman et al., 2001, 2013). The broad trends and magnitude of precipitation are well represented in the model. The average daily precipitation over South America in Phase I is significantly lower than in Phase II and largely concentrated in the North-West. In Phase II, the average rate is much higher and the precipitation spreads much further into the central states. However, some fine detail is missed in the model and the precipitation does not spread as far east as the TRMM data suggests. For example, there are several instances of storms in phase II between 45 and 50° W not reproduced in the model.

Precipitation trends over the course of the campaign had a strong impact on the BBA concentrations in the western regions, both because increased precipitation reduced the number of fires and increased the level of wet deposition in the biomass burning regions. Phase I was characterised by the accumulation of regional haze, with some localised removal events. Widespread precipitation throughout Phase II largely washed out the accumulated haze, but continued burning maintained

a polluted haze, albeit relatively clean compared to Phase I. Throughout, conditions remained dry in the Eastern states.

Drop-sondes were used during the SAMBBA flights to measure temperature, moisture content and wind speed in the atmospheric column. Skew-T plots from drop-sondes from four flights are compared with model data in Fig. 4. Skew-T plots for all other drop-sondes made during the SAMBBA campaign can be seen in the Supplement. The model generally represents the coarse structure and wind direction of the column well. However it fails to reproduce some of the fine detail. This is unsurprising given the relatively coarse vertical and horizontal resolution of the model. The fit for the temperature profile is better than for the dewpoint profile, with several examples of stratification in the dewpoint profile observed in the flights not seen in the model. For example between 850 and 700 hPa in flight B737 (Fig. 4C), the model significantly overestimates the moisture content of the atmosphere. It was observed on the SAMBBA flights that these dew point inversions would cap aerosol transport, forming distinct layers. This is a phenomena we are unlikely to reproduce in the model. The top of the modelled boundary layer, inferred from the lowest inversion in the temperature profile, is generally close to that observed in the measurements, but not as clearly defined or strong.

5.2 Horizontal distribution and optical properties of aerosol – comparison with remote sensing data

Figure 5 shows averaged aerosol optical depth (AOD) at 550 nm over the two phases of the campaign. The panels on the left show AOD from combined MODIS ~~and TERRA~~ Aqua and Terra satellites, whilst the centre and right panels show AOD from model runs using standard 3BEM emissions and the modified emission setup respectively.

Phase I is characterised by a build up of BBA, forming a large regional haze with high AOD over much of central South America. The magnitude of the AOD is well captured in the model, and is closest to that observed by the satellites in the modified emission scenario. However, the distribution is displaced: the highest AODs observed by the satellites are in central Mato Grosso state, around 55° W and 15° S, while in both model runs it is in Rondônia state further to the north west, particularly about a cluster of fires at 64° W and 10° S. This is location of greatest fire emissions in both emission products, as shown in Figure 2. As this does not show as strongly in the satellite data, emissions are presumably too strong at this location.

During Phase I, both model runs ~~also~~ show a significant proportion of BBA ~~are transported west~~ transported west not observed by the satellite AOD measurements in Figure 5. This is due to a combination of both a greater proportion of the emissions originating in western states/forest ~~fires~~ biomes and a greater proportion of the aerosol being in the upper levels of the troposphere. Figure 4a and b show easterly winds in the free troposphere and northerlies in the boundary layer over these flights.

During Phase II, both model and satellite data show reduced AOD over much of the domain. The satellite measurements show a large reduction in BBA over Rondônia, but significant AOD in

the North-Eastern states where most fires are cerrado. In the model runs, there is an eastward shift compared to Phase I, particularly in the modified emission scenario, but AOD in the eastern regions is still lower than that observed by the satellites. ~~In addition there are significant regions of high modeled AOD in the western states not observed by the satellites.~~ Mean, standard deviation and spatial correlation coefficients of AOD for Phases I and II are given in Table 6. Compared to the standard 3BEM emissions scenario the modified emission scenario shows higher mean AOD in both Phases, stronger correlation in Phase I, but weaker correlation in Phase II.

Figure 6 shows the timeseries of AOD at 550 nm measured at 4 of the AERONET sites marked in Fig. 5, including measurements from overpasses of the MODIS ~~AQUA and TERRA~~ Aqua and Terra satellites. The panels on the left show the standard 3BEM emissions and the panels on the right are for modified emissions. There is little difference in AOD simulated at these sites between the two emission scenarios. With the exception on the Cuiaba site, the model replicates the build up of aerosol and AODs in the first half of the campaign well (although it should be noted that f_x was tuned to be able to represent the magnitude of AODs in this part of the campaign). The Cuiaba site is likely too low in the model because this region is more dominated by cerrado fires, whereas the other sites have a greater proportion of forest fires nearby.

In Phase II of the campaign, the model runs overestimate the AOD over every Aeronet site evaluated against. It proved to be a challenge to find a suitable scaling factor to enable a large enough build up of AOD in the first half of the campaign without "overshooting" in phase II. This may be due to the model not washing out aerosol as efficiently as it should, the emissions not decreasing in intensity enough in the second half, or a combination of these factors.

5.3 Comparisons with in-situ aircraft measurements

5.3.1 Vertical distribution of CO and BBA

In this section of the paper, we will be comparing model results with in-situ measurements of aerosol and aerosol optical properties from flights conducted during the SAMBBA campaign. The remainder of the analysis will focus on four flights as case studies: B731, B734, B739 and B742 on 14, 18, 23 and 27 September 2012 respectively. The ~~flight details are~~ instrument coverage of these flights is summarised in Table 4. These flights were selected as they extensively sampled the regional haze across the range of environments and meteorological conditions encountered during the campaign, with near complete instrument coverage. Flights B731, B734, B739 sampled the regional haze in Rondônia state, characterised by cleared and pristine forest, whilst B742 sampled over Tocantins state in the Cerrado (savannah-like) environment. All aerosol data from the model has been summed over bins where D_p is $< 1\mu\mathrm{m}$ (defined as all bins 1–4 and 67.8 % of bin 5) and converted to standard temperature and pressure units ($\mu\mathrm{g\,sm}^{-3}$) for comparison with submicron flight measurements.

The paths of the flights used in this study are shown in Fig. 7. Following a profile ascent out of the host airport (Porto Velho for B731, B734 and B739, Palmas for B742), the aircraft travelled to the region of interest at high altitude (7–8 km a.s.l.), before descending to near surface via a stack of straight and level runs at altitudes above and within the boundary layer. Flight B739 was a slight exception to this pattern, with only a brief period at high altitude, and without the straight and level runs in the stacked formation. Near surface, flights B739 and B742 sampled extensive small plumes in the area, resulting in non-uniform flight patterns. All flights then returned either at high altitude (B731, B734) or high within the boundary layer (B739, B742), before profile descent back to base. Each flight therefore had a number of profiles and straight and level runs at multiple altitudes, providing a comprehensive characterisation of the haze in the region sampled. The boxes around each of the flight paths in Fig. 7 show the area averaged over when calculating the statistics from the model when carrying out the comparisons.

Figure 8 shows vertical profiles of CO, POM and scattering coefficient at 550 nm (b_{scat}). CO is used as a relatively inert tracer, largely unaffected by precipitation or wash-out. POM is shown and compared with AMS organics data as it makes up the dominant fraction of the total aerosol budget. Finally, b_{scat} is used to show the optical depth of the aerosol. b_{scat} is used rather than b_{ext} to avoid additional measurement uncertainty by the addition of b_{abs} (Bond et al., 2013). ~~The dashed blue lines and shaded regions show median, interquartile and 5th–95th percentile range derived from the standard 3BEM emission scenario, while the red lines and shaded regions are for the modified emission scenario. The solid black lines show median values from the profiles conducted by each flight , while the fine grey lines show the actual flight track data . The flight data~~ The flight data is limited by never flying above 8 km altitude. However, as a significant portion of the plume-rise emissions in the standard 3BEM case are emitted above 8 km (see Fig. 2), the profiles from the model runs are plotted up to 12 km. This measurement evaluation is an improvement over Longo et al. (2010), where the plume-risen emissions were compared against flights which did not fly above 4 km and comparisons were only made with CO.

B731 coincided with the end of a long build up of aerosol in Rondônia before it was washed out during the progression into the wet season and had some of the highest measurements of aerosol in the campaign. Both model scenarios under-predict CO and POM within the boundary layer and over-predict above the boundary layer. The flights show the majority of CO and aerosol are in the lower 2 km of the troposphere, with a steep drop off above this. Both model runs show a secondary peak in aerosol above the boundary layer, between 4–5 km in the updated emissions scenario and around 7 km using the standard 3BEM emissions. In both model runs, too large a proportion of the emissions are being emitted above the boundary layer. The same elevated peak can be observed in b_{scat}, although it decreases faster above the boundary layer than POM. This is because POM is in units at standard temperature and pressure and independent of altitude, while b_{scat} is related to the

absolute density of particles and decreases exponentially with altitude. b_{scat} is therefore dominated by aerosol in the boundary layer in both flight and model.

By the time of flight B734, significant precipitation had occurred over Rondônia, reducing the aerosol loadings in both model and measurements. The flight is also sampling a different region of Rondônia. CO in the boundary layer is also lower, implying reduced fire emissions. Below 4 km, flight CO and POM are similar to the modified emissions scenario. Above 4 km, CO remains elevated in both measurement and model. POM sharply decreases in the flight data, while in the model it is clear the POM has been emitted at the same height as the CO and follows a similar profile. The lack of observed POM at the same altitude as CO implies either the wash-out processes are not being well represented in the model, both CO and POM are being emitted at altitude in an unrealistic fashion with less of a negative impact on CO or the flight is measuring a source of CO that does not have much associated POM.

Flight B739 was conducted at the start of Phase II, by which time the majority of accumulated aerosol in the western states had been washed out. During this flight, there were large stratocumulus clouds and significant convection over the region. The increased soil moisture after previous days precipitation ~~meant many of the fires were smouldering~~ resulted in a larger fraction of smouldering fires. Given the limitations of the model setup, we would expect this flight to be the most challenging of the case studies for the model. High concentrations of CO and slightly elevated POM in the lowest km of the boundary layer are observed, but these fresh emissions have not become well mixed at the time of flights. Aside from that, the measured atmosphere is relatively clean compared to the earlier flights. The standard 3BEM emission scenarios is close to the measurements for CO, at least up to 6 km altitude, whereas the modified emission scenario has too much CO. However, both model scenarios ~~overpredict~~ over-predict POM in and above the boundary layer. The elevated peak in POM and CO in the model is much higher during this period, especially in the standard 3BEM case where it is above where the flights can observe. While the existence of this layer cannot be ruled out, from the good agreement between aircraft and satellite derived AOD it can be inferred that the magnitude of aerosol loadings are unlikely (see Darbyshire et al., ~~2014~~in prep.). This elevated peak results from a combination of high plume-risen injected emissions and convective transport.

Flight B742 was carried out in the eastern Tocantins state. This region is dominated by ~~cerrado~~ Cerrado fires. It is clear that the magnitude of emissions are too low in the region. CO, POM and b_{scat} are higher in the modified emissions scenario, but still approximately 50 % below measured. However, the shape of the vertical profile is well represented, with flights and both model scenarios showing aerosol and CO well mixed within the boundary layer, and little above it. The lower carbon density of the cerrado biome to tropical forests results in less intense fires, with the injection height ~~is~~ rarely much higher than the top of the boundary layer.

Overall, flight B734 shows the closest correspondence between the measurements and model data of the case studies. The modified emissions do produce on average a more reasonable injection height

to represent flaming emissions. However, there is still a strong bias towards overestimating the injection height, particularly over tropical forest biomes. This is most apparent in POM~~, while modelled~~. Modelled CO may be similar to flights even where POM diverges. b_{scat} decreases exponentially with altitude, meaning the high altitude layers are optically thinner than those in the boundary layer. However, this may still be a significant divergence from reality, given the negligible measured b_{scat} at these heights.

5.3.2 Composition, optical properties and size distribution of aerosol

Box and whisker plots of BC, POM : BC ratio and single scattering albedo (ω_0) for the straight level runs below 3 km of the atmosphere are shown in Fig. 9. ~~During~~ The SP2 had insufficient coverage during flight B731 ~~, the SP2 was not functional for much of the flight and had little crossover with when the AMS was working, and so has been left out of this section of the analysis, although~~ to provide POM:BC ratios, hence these are not included here. However, ω_0 measurements for ~~B731 have been included in the Supplement~~this flight are presented in the supplement. Model data is from the modified emissions scenario, extracted along the flight path by finding the x-y grid point closest to the flight measurement, then linearly interpolating in the vertical and time to the altitude and time of reading. There was little difference in composition between the two scenarios.

The western flights show a higher POM : BC ratio on average compared to the Eastern flight B742. In both western flights, the modelled POM : BC ratio is much lower than measured, due to the increased loadings of BC. The modelled POM : BC ratio is consistently between 9 and 11, slightly higher on B739 and lower in B742. The median measured ratio for B734 is 14.5 and for B739 it is 17.6. B739 is likely higher due to the increased proportion of smouldering fires post precipitation, which tend to have higher POM : BC ratio. In the eastern flight B742, the median POM : BC ratio is 9.1, similar to the modelled, although the range is still larger. The lower POM : BC ratio in flight B742 is likely due to the higher proportion of cerrado fires.

The POM : BC ratio shows a lot more variability in the flight data compared to the model. The variation is likely due to a combination of varying emission factors (EF) due to fuel type, flaming temperature, burning efficiency, and other factors (Jolleys et al., 2012); and SOA formation (Jimenez et al., 2009). The model emissions do not vary in composition to the same extent, due to limited measurements driving the Andreae and Merlet (2001) EF, and no SOA formation is represented in the MOSAIC mechanism. Some recent measurements, such as Jolleys et al. (2012), suggest that, unlike urban plumes, there is little net SOA formation during the ageing of BB plumes, supporting the primary OC assumption in heavily BB influenced regions. However, other studies, such as Vakkari et al. (2014), suggest growth by SOA condensation in the first few hours of plume ageing is a significant factor in determining BBA composition.

Modelled ω_0 is largely controlled by the ratio of BC to other aerosol components. In flights B734 and B739, the flight average is similar to modelled ω_0~~, if slightly higher on average~~. B739 shows

a much greater degree of variability, with an ~~IQR of 0.86~~interquartile range (IQR) of 0.81–0.95. However, it should be noted that the PSAP instrument had only partial coverage during this flight, which may be skewing some of the data. ~~However, while~~ While the POM : BC ratio is always lower in the model, ω_0 is often lower in the measurements. Given the low modelled POM : BC ratio, the model should be underestimating ω_0 by a similar margin; i.e. it is getting ω_0 right for the wrong reasons. In contrast, flight B742 has a similar POM : BC ratio between flight and model but significantly lower ω_0 (the model is getting it wrong for the right reasons). The implication is that there are properties of the aerosol affecting how it absorbs radiation not being captured in the model. The mixing rule (in this case Maxwell-Garnett) may be under-predicting the absorption amplification of the other aerosol components and/or the organic portion of the aerosol should be slightly absorbing in the visible spectrum ("brown" carbon). ~~Explicit resolution of the aerosol mixing state , as is done by could also improve results~~In addition, recent WRF-Chem developments have enabled explicit modelling of the mixing state of BC with other components (Matsui et al., 2013). While more expensive to run, using this method may improve predictions of aerosol absorption.

Figure 10 shows the CCN concentration~~and size distribution~~ , number and volume distributions of aerosol from flights B734 and B742 compared with the modified emission scenario. Data was extracted from the model along the flight path. In both flights, the peak in the size distribution is the same (within error), showing the studies the modelled distribution is based on are representative of regional BBA. However, the ~~modeled~~ modelled distribution is too wide, with too much aerosol ~~is~~ in the larger bins between 1 and 5 µm and too little in the accumulation mode. This implies that there is too much emitted coarse mode BBA, there is another source of coarse aerosol (e.g. dust) in the model not observed in the flight, too much coarse aerosol is being transported up to flight height, or the process of larger BBA particles being preferentially removed by precipitation (as Taylor et al., 2014, show with Canadian fires) is not being well captured in the model. However, it should be noted that the GRIMM data has some minor uncertainties attributed to it due to line-losses and refractive index ~~. The results presented~~ and the results presented here should be seen as a lower limit. Further sensitivity work is needed to test which of these factors are more important. The model represents the spread of CCN well in flight B734, with the measured CCN at ~~0.14~~between 0.135 and 0.154 % supersaturation ~~($CCN_{0.14}$)~~ in between the ~~modeled~~ modelled $CCN_{0.1}$ and $CCN_{0.2}$ values. The model also underestimates CCN concentrations over flight B742, in line with the under-prediction of aerosol loadings over the eastern regions.

6 Conclusions

We have modified the online ~~couple~~ coupled regional model WRF-Chem to use 3BEM emissions and plume-rise parameterisation with the MOSAIC sectional aerosol and CBM-Z gas phase chemistry mechanisms. The default values of both active fire size and burned area given in PREP-CHEM-SRC

are 22.8 ha (Longo et al., 2010). Using these values it was found that the injection height ~~would often be~~ was often biased high. Given the downward trend in fire sizes in Brazil from 2000, emissions suitable for the 2012 Brazilian biomass burning season ~~have been developed . Estimates are~~ were developed using estimates based on FRP measurements over the ~~2012 South American biomass burning season~~region, with different values ~~used~~ for different biomes. In the modified inventory, burned area and active fire size are treated independently, ~~where the burned area is used to calculated~~ with burned area used to calculate the emitted mass and active fire size to calculate the injection height of the plume-rise parameterisation. Results from ~~the~~ model simulations have been compared against in-situ measurements from the SAMBBA flight campaign.

In many modelling studies~~and emission products~~, an enhancement factor (f_x) is required to scale fire emissions to produce reasonable AODs (e.g. Wu et al., 2011a; Kaiser et al., 2012; Tosca et al., 2013). The need for f_x highlights the many uncertainties in calculating biomass burning emissions (Ichoku et al., 2012). ~~Factors causing this include underestimating the biomass density or burn fraction, satellite products missing some fires, due to overpass times, cloud cover, fires being too small to detect, or multiple fires within the same 1being assigned a single fire pixel.~~ In this study, we found when ~~updating~~ modified the estimated burned area for 2012 values, the total emitted mass was significantly smaller. We therefore increased ~~the emission amplification factor~~ f_x from 1.3 to 5 ~~in order~~ to produce reasonable AODs within the model. The implication is that using the standard 3BEM emission product the modelled AOD was reasonable, but only because the burned area was larger than the 2012 season average. Using our best estimate of burned area required a scaling of emissions to compensate.

In the western regions over the first half of the campaign, modelled AODs compared well to satellite measurements. However, ~~in the second half of the campaign the model consistently overestimate AODs in the western regions. Over Eastern~~ AODs were consistently overestimated in the second part, when there was more precipitation. Over eastern cerrado regions, the model underestimated AOD over the whole campaign. There are several factors that may explain these observations. ~~We used an average burned area across the whole season, which does not vary if fires were smaller and less vigorous in the second phase~~ Firstly, the average burned areas used in the study did not vary over the course of the campaign. ~~Distribution of fire size is heavily skewed to mostly small fireswith a few massive ones, something not represented using a mean fire size~~Secondly, small fires are often not detected. Randerson et al. (2012) estimate some 35 % of fire emissions are missed globally due to lack of detection of small fires, with this factor being larger in some regions. Observations on the SAMBBA flights were that in some regions there were more than one fire per km^2, particularly in the eastern cerrado burning states~~. estimate some 35of fire emissions are missed globally due to lack of detection of small fires, with this factor being larger in some regions. The emissions inventory of~~ , which would be identified as a single fire. Finally, the Yevich and Logan (2003) ~~has been used in this study~~ inventory was used to account for small ~~scale~~ biofuel and agricultural burnings. However,

~~the~~ this inventory provides annual averages for emissions which are known to show large seasonal variability (Duncan, 2003). Adding a function to control the seasonal variation ~~in these emissions would~~ of these emissions and increase their contribution ~~to the aerosol loadings~~ in the dry season ~~, which~~ should provide better estimates, particularly ~~in rural~~ over agricultural areas such the eastern cerrado states.

Over the western flights, which were dominated by tropical forest fires (and pasture burnings), there was too much emitted mass at high altitude in both model scenarios. With fire size significantly smaller in the modified emission scenario, the injection height was typically 2–3 km lower, but still approximately 2 km above the boundary layer. ~~show that the majority of tropical forest fires inject into the boundary layer, with only a few large outliers penetrating higher.~~ The distribution of fire size is positively skewed, with the majority of fires being small ($\ll 5\,\mathrm{ha}$) and only a few large fires (some $50\,\mathrm{ha}$ or larger). ~~Using a simple average does not represent this distribution. A better probabilistic representation~~ A probabilistic representation of this distribution may be needed~~to represent the size distribution of fires in the model to account for this.~~ .

The vertical stability in the atmospheric column from the model was compared with dropsonde measurements from the ~~flight~~flights. The coarse structure was well captured but much of the fine detail was not~~represented in the model.~~ . The model failed to reproduce the temperature and dewpoint inversions at the top of the boundary layer, likely due to vertical resolution issues and limitations of the PBL parameterisation. The stability profile from the parent model is used to define the column of the plume rise parameterisation. Without a clearly defined stable layer~~in the temperature profile~~, it is ~~unsurprising the parameterisation~~ perhaps unsurprising that it often penetrates the ~~boundary layer~~PBL. Forcing a small temperature inversion at the PBL top ~~in~~ may improve the plume-rise parameterisation~~may be needed to improve its accuracy~~'s accuracy, but day to day and geographical variability makes such an intervention impossible without comparison of the hindcast with measured data.

The vertical distribution of carbon monoxide (CO), particulate organic matter (POM) and scattering coefficient (b_{scat}) were compared between model runs and flight measurements. The modelled CO vertical profile was reasonably well represented, as seen in previous studies (Freitas et al., 2007, 2009; Longo et al., 2010). However, there were regions of elevated aerosol layers in the model not observed in flight measurements. Aerosol has many more loss processes than CO, particularly through wash-out. Andreae et al. (2001) show convective transport of tropical BBA is important for forming aerosol layers at high altitude. However, only around 5–20 % of accumulation mode aerosol is retained during transport; the rest is washed out. The plume-rise parameterisation transports 100 % of flaming emissions when convection is triggered. Accounting for the aerosol loss processes attributed to convection during plume-rise may be needed to better represent the aerosol profile.

The model failed to represent the same variation in aerosol composition and ω_0 observed in the flights. This composition in the model is driven by the Andreae and Merlet (2001) emission

factors (EF). Akagi et al. (2011) have reviewed many more recent studies to provide newer estimates. The OC : BC ratio for savannah has remained the same at 7.08. However, the estimated tropical forest EF increased from 7.88 to 9.05, approximately 15 % higher. Using these updated EF would bring the model closer to ~~typical~~ measured POM : BC ratios in the western flights. Work is underway to update the PREP-CHEM-SRC to the EF of Akagi et al. (2011). Representing flight B739 will still be a challenge however, given the impact of precipitation on fire conditions. ~~Using different EF for smouldering and flaming emissions, with flaming and smouldering fraction varying dynamically withsoil moisture, may be able to represent this variation of emissions.~~ This may be accounted for using dynamic EF varying with, for example, soil moisture. More detailed measurements would need to be collected and reviewed to develop an emissions inventory with this flexibility. It should also be noted that comparisons are between modelled primary organic matter ~~from the model with~~ and measured total organic matter ~~from measurements (including contribution from SOA). Developments including SOA treatment in~~ (including SOA mass). Work is being conducted to run WRF-Chem ~~, such as using the VBS , could be needed to represent the observed variation in composition~~with a Volatility Basis Set (VBS; Donahue et al., 2011; Shrivastava et al., 2011) over the SAMBBA period to simulate SOA formation and enable more in depth aerosol compositional comparisons with flight cToF-AMS data.

~~Modeled~~ Modelled ω_0 was often too high when the POM : BC ratio was approximately correct, and close to measured when POM : BC ratio was too low. ~~Improving the aerosol composition in the model is needed before we can evaluate the ω_0. However, this~~ This indicates failure of the model to accurately predict the aerosol optical properties from the composition. ~~The behaviour of the optical calculations can be tested by initialising the optical properties subroutine with SAMBBA flight measurements, showing how much of the discrepancy is due to inadequacies in the calculations . The~~ The model may be underestimating the enhancement factor of BC and a better mixing-rule is needed (such as shell-core), or explicit modeling of the BC mixing state (Matsui et al., 2013). Some SW absorption due to the "brown carbon" components of organic aerosol is also likely needed (Lack et al., 2012, 2013; Saleh et al., 2014). The discrepancies highlight the need to capture the full mixing state, including both SOA and POA, as well as condensable inorganic vapours, to ~~represent~~ accurately predict aerosol optical properties.

The model represented size distribution peak location well in flights B734 and B742. CCN concentrations correspond well over the western flight B734, with $CCN_{0.2}$ between 900 and 1100 scm^{-3} within the boundary layer. Over the eastern flight, the model ~~underpredicted~~ under-predicted CCN concentration. However, the low CCN concentrations are in line with the low aerosol loadings over this flight and period.

Appendix A: MACC-II Boundary Conditions

Only a subset of chemical species thought to be significant in long-range transport and chemistry are included in the MACC-II product: CO, O_3, OH, SO_2, NO_2, HNO_3, CH_4, C_2H_6, isoprene, peroxyacetyl nitrate (PAN) and formaldehyde (HCHO). The aerosol module used in MACC-II is described by Morcrette et al. (2009). Five species of aerosol are carried: natural sea salt (SU) and dust (DU), and three anthropogenic aerosol (POM, BC and SULF). SULF, POM and BC are each treated as bulk aerosol, with BC and POM treated as two components – hydrophobic and hygrophilic. SS and DU are each represented by bins with boundaries at 0.03, 0.5, 5 and 20 µm diameter for SS and 0.03, 0.55, 0.9 and 20 µm for DU (Morcrette et al., 2009).

The model uses log-normal distributions with parameters of mean diameter (D_p) and geometric standard deviation (σ) as defined below (Jean-Jaques Morcrette, personal communication, 2013):

- SS: two log-normal distributions; the first with $D_{p,1} = 0.389$ µm, $\sigma_{p,1} = 1.9$, $N_{\mathrm{tot},1} = 70$, the second with $D_{p,2} = 3.984$ µm, $\sigma_{p,2} = 2.0$, $N_{\mathrm{tot},2} = 3$.
- DU: a single log-normal distribution, $D_p = 0.58$ µm, $\sigma_p = 2.0$.
- The bulk aerosol BC, POM and SO_4^{2-} is assumed to be in an accumulation mode with single log-normal distribution, D_p=0.071 µm, σ_p=2.0.

The fraction of each MACC-II bin to be partitioned into each MOSAIC bin is given by the fraction of each distribution that falls between each MOSAIC bin boundary. As the upper limit of MOSAIC aerosol is 10 µm, all aerosol mass from the distributions above 10 µm is discarded. See Table 3 for full apportionment to each MOSAIC size bin.

The SULF carried in MACC-II is assumed to be ammonium sulphate ($(NH_4)_2SO_4$) when mapped to the WRF-Chem MOSAIC species, in order for the aerosol to have neutral acidity. Likewise, SS is assumed to be NaCl and is split between the Na^+ and Cl^- ions. The MACC-II boundary conditions were interpolated to the model grid using a modified version of the mozbc script (www.acd.ucar.edu/wrf-chem).

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Table 1. ~~Table~~ Fractional apportionment of ~~fire area and~~ particulate emissions across the 8 MOSAIC size bins, ~~derived from MODIS FRPmeasurements~~ showing range of particle diameters for ~~the 2012 Brazilian fire season~~each bin, ~~prepared by G~~primary anthropogenic emission size fraction and biomass burning emission fractions based on Janhäll et al. (2010).~~Periera.~~

Bin 1	Bin 2	Bin 3	Bin 4	Bin 5	Bin 6	Bin 7	Bin 8
Particle dry diameter (nm).							
39.1–78.1	78.1–156	156–313	313–625	625–1250	1250–2500	2500–5000	5000–10000
Primary anthropogenic aerosol emission size fractions (fine mode, $< 2.5\,\mu m$).							
0.06	0.045	0.245	0.40	0.10	0.15	0.0	0.0
Biomass burning aerosol emission size fractions, based on Janhäll et al. (2010).							
0.0092	0.1385	0.4548	0.3388	0.0567	0.0020	0.0	0.0

Table 2. Table of fire area and size, derived from MODIS FRP measurements for the 2012 Brazilian fire season.

Biome	number of ~~data~~ data points	Burned area A_{fire} [ha]	Active fire size S_{fire} [ha]	Ratio ~~(S/A)~~ (S_{fire}/A_{fire})
Forest	191 386	4.3 ± 8.3	1.15 ± 2.30	0.267
Mixed Forest	1756	10.63 ± 12.16	2.45 ± 3.01	0.305
Scrublands	95 681	9.13 ± 12.0	2.15 ± 2.30	0.235
Savanna/cerrado	226 493	7.80 ± 9.30	1.90 ± 3.20	0.244
Cropland	36 667	9.72 ± 10.4	1.33 ± 2.46	0.137

~~Fractional apportionment of particulate emissions across the 8 MOSAIC size bins, showing range of particle diameters for each bin, primary anthropogenic emission size fraction and biomass burning emission fractions based on . Bin 1 Bin 2 Bin 3 Bin 4 Bin 5 Bin 6 Bin 7 Bin 8 39.1–78.1 78.1–156 156–313 313–625 625–1250 1250–2500 2500–5000 5000–10 000 0.06 0.045 0.245 0.40 0.10 0.15 0.0 0.0 0.0092 0.1385 0.4548 0.3388 0.0567 0.0020 0.0 0.0~~

Table 3. Fractional apportionment of aerosol loadings from MACC-II model to 8 MOSAIC size bins for initial and boundary conditions (Morcrette et al., 2009). Apportioning for MACC-II aerosol species black carbon (BC), organic aerosol (OA), sulphate aerosol (SULF), dust (DU) and sea salt (SS). Uses same MOSAIC dry particle diameters as Table 1.

Bin 1	Bin 2	Bin 3	Bin 4	Bin 5	Bin 6	Bin 7	Bin 8
BC, POM (hydrophobic and hygrophilic) and SULF.							
0.0246	0.1475	0.3506	0.3321	0.1253	0.0187	1.1×10^{-3}	2.4×10^{-5}
SS Bin 1: 0.03–0.5 µm.							
1.1×10^{-3}	0.0312	0.3169	0.6502	0.0	0.0	0.0	0.0
SS Bin 2: 0.5–5.0 µm.							
0.0	0.0	0.0	0.01	0.04	0.164	0.786	0.0
SS Bin 3: 5.0–20 µm.							
0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.5515
DU Bin 1: 0.03–0.5 µm.							
2.1×10^{-5}	0.0023	0.0928	0.9049	0.0	0.0	0.0	0.0
DU Bin 2: 0.55–0.9 µm.							
0.0	0.0	0.0	0.1493	0.8507	0.0	0.0	0.0
DU Bin 3: 0.9–20 µm.							
0.0	0.0	0.0	0.0	0.0989	0.3736	0.3643	0.1415

Table 4. ~~Summary~~ Table of ~~physical parameterisations~~ instrumentation used during SAMBBA flights B731 (14 September 2012), B734 (18 September 2012), B739 (23 September 2012) and B742 (27 September 2012). The coverage of each instrument for each flight is indicated by the categories: Full ($> 80\%$ coverage), Partial (between 80% and 30%) or Insufficient ($< 30\%$). For details of instruments see text in ~~WRF-Chem model runs~~Section 4.2. Mass and number mixing ratios given per unit volume at standard temperature and pressure (sm^{-3} or scm^{-3}).

~~Process WRF-Chem Option Reference Microphysics Morrison 2-moment Aerosol Activation Abdul-Razzak and Ghan Cumulus parameterisation Grell 3-D Planetary Boundary Layer Yonsai University (YSU) Surface Layer MM5 surface-layer similarity Land-Surface Model Unified NOAH land-surface Longwave Radiation RRTMG Shortwave Radiation RRTMG~~

~~Table of instruments and flights used for model evaluation. Each flight shown to have Full-, Partial- or Insufficient-coverage for each instrument, where Full is $>80\%$ coverage, partial is between 80% and 30%, and Insufficient is $<30\%$. Acronyms used for instruments: Single Particle Soot Photometer , compact Time of Flight Aerosol Mass Spectrometer , Aero-Laser AL5002 VUV resonance fluorescence gas analyser, 3-wavelength integrating nephelometer , Particle Soot Absorption Photometer (PSAP) , Scanning Mobility Particle Sizer , a GRIMM model 1.108 Optical Particle Counter and a DMT dual column Cloud Condensation Nuclei counter (CCNc) . Mass and number mixing ratios given per unit volume at standard temperature and pressure (or).~~

Instrument	Measurement	Units	Temporal resolution	B731	B734	B739	B742
SP2	BC	$\mu\mathrm{g\,sm}^{-3}$	1 s	Insufficient	Full	Full	Full
cToF-AMS	POM	$\mu\mathrm{g\,sm}^{-3}$	$\approx$ 30 s in level runs $\approx$ 10 s during profiles	Partial	Full	Full	Full
AL5002 VUV	CO	ppbv	1 s	Full	Full	Full	Full
Dry Nephelometer	b_{scat}	km^{-1}	1 s	Partial	Full	Full	Full
PSAP	b_{abs}	km^{-1}	25-30 s	Partial	Full	Partial	Partial
SMPS	Number distribution (20–350 nm)	scm^{-3}	$\approx$ 60 s	Partial	Full	Insufficient	Full
GRIMM	Number distribution (0.3–20 µm)	scm^{-3}	$\approx$ 6 s	Full	Full	Full	Full
CCNc	CCN Concentration	scm^{-3}	1 s	Full	Full	Insufficient	Full

Table 5. Summary of physical parameterisations used in WRF-Chem model runs.

Process	WRF-Chem Option	Reference
Microphysics	Morrison 2-moment	Morrison et al. (2005)
Aerosol Activation	Abdul-Razzak and Ghan	Abdul-Razzak and Ghan (2002)
Cumulus parameterisation	Grell 3-D	Grell and Devenyi (2002)
Planetary Boundary Layer	Yonsai University (YSU)	Hong et al. (2006)
Surface Layer	MM5 surface-layer similarity	Zhang and Anthes (1982)
Land-Surface Model	Unified NOAH land-surface	Ek et al. (2003)
Longwave Radiation	RRTMG	Mlawer et al. (1997)
Shortwave Radiation	RRTMG	Pincus et al. (2003)

Table 6. Table of mean, spatial standard deviation and centred Pearson's product-moment correlation coefficient; comparing AOD at 550 nm from the two WRF-Chem emissions scenarios with the combined MODIS Terra and Aqua satellite data. Data used same as to plot Figure 5.

Dataset	Phase I			Phase II		
	Mean	Standard deviation	Correlation coefficient	Mean	Standard deviation	Correlation coefficient
MODIS	0.321	0.190	N/A	0.221	0.131	N/A
Standard 3BEM	0.355	0.129	0.678	0.285	0.117	0.623
Modified 3BEM	0.381	0.155	0.732	0.286	0.131	0.591

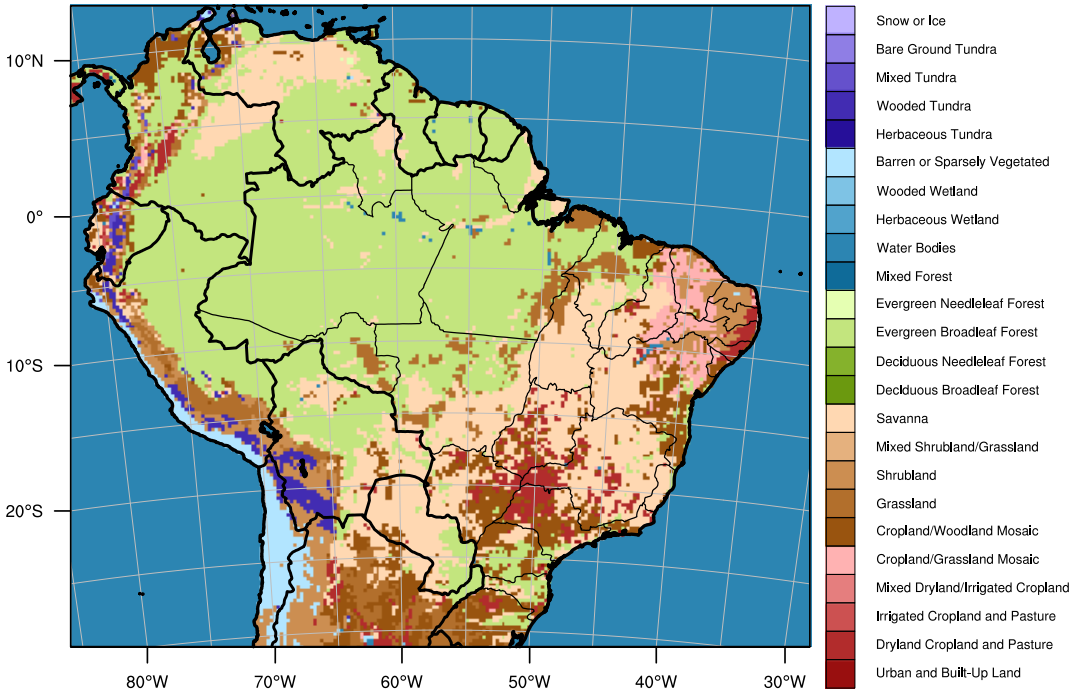

Figure 1. Map of domain used for study, at 25 km horizontal grid spacing with lambert projection. Coloured by 24 USGS land-use categories. The southern Amazon, coloured green, is the main region of deforestation burning, corresponding to the West-central Brazilian states and northern Bolivia. The East-central Brazilian states, coloured pale-brown, are the main regions of cerrado burning.

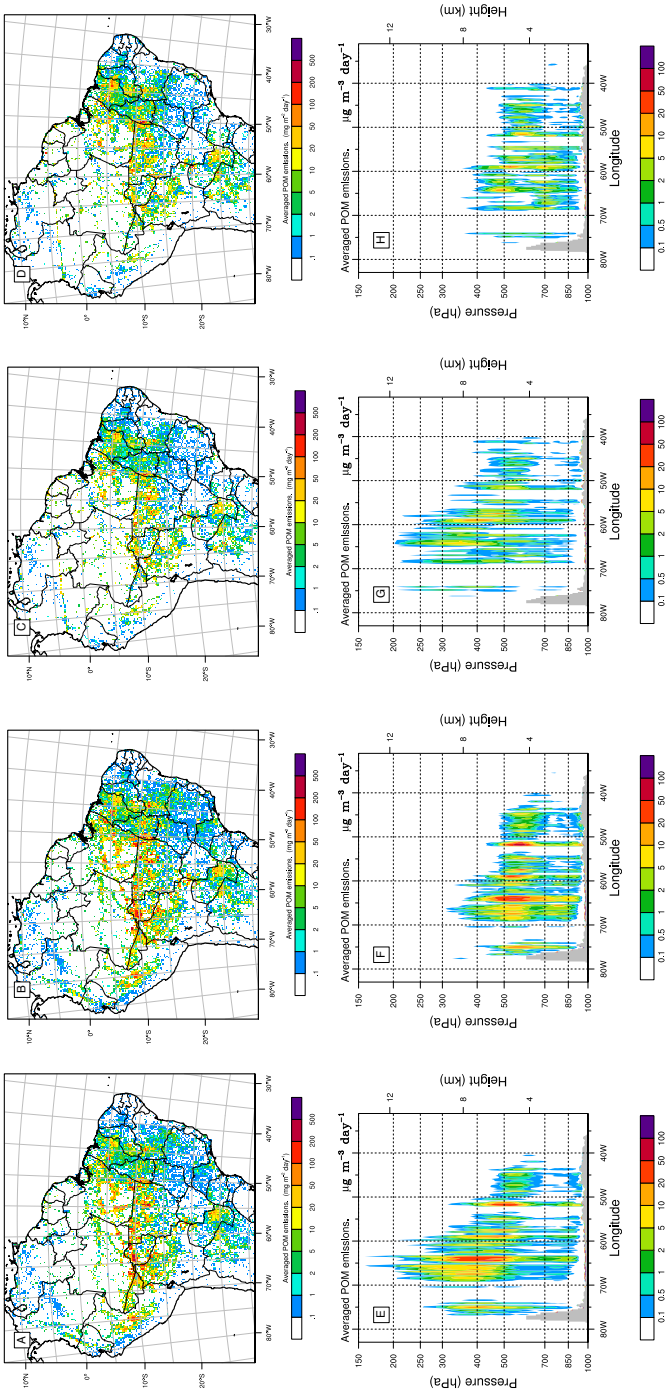

Figure 2. Emissions of organic aerosol (OA) over the course of the campaign. Panels **(a–d)** are maps of emissions, showing total emissions in the atmospheric column ($\mathrm{mg\,m^{-2}\,day^{-1}}$). Panels **(e–h)** are vertical profiles of emissions through a transect along 9 ° S ($\mathrm{\mu g\,m^{-3}\,day^{-1}}$). Panels **(a)**, **(b)**, **(e)** and **(f)** show averaged emissions over Phase I of the campaign (6–22 September 2012). **(c)**, **(d)**, **(g)** and **(h)** are averaged over Phase II (23–30 September). Panels **(a)**, **(c)**, **(e)** and **(g)** are for the traditional 3BEM emissions. Panels **(b)**, **(d)**, **(f)** and **(h)** are for the modified emissions, using smaller fire size and burned area depending on vegetation type as described in Table 2.

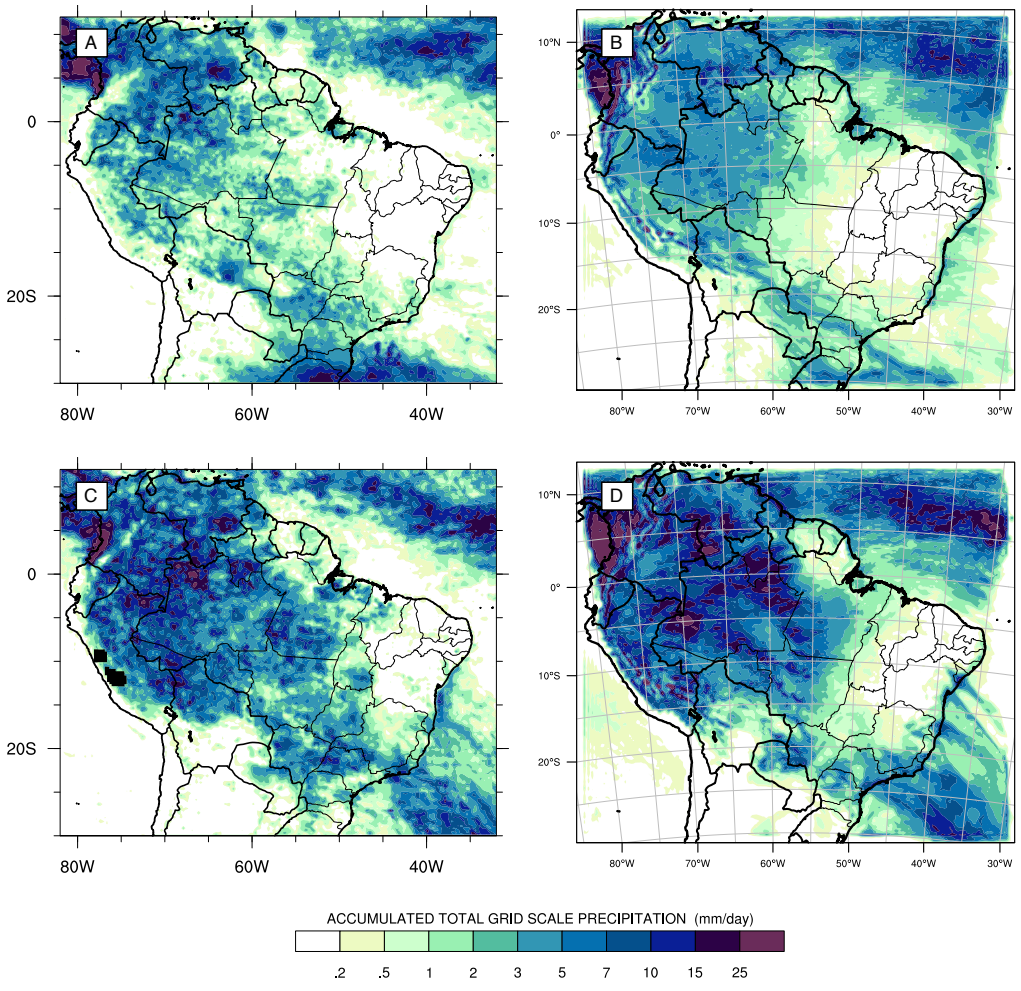

Figure 3. Maps of averaged precipitation ($\mathrm{mm\,day^{-1}}$). **(a** and **c)** are derived from the TRMM 3B42 satellite product (Huffman et al., 2001, 2013). **(b** and ~~**c)**~~ **d)** from WRF-Chem model runs. **(a** and **b)** for Phase I (6–22 September 2012), **(c** and **d)** over Phase II (23–30 September).

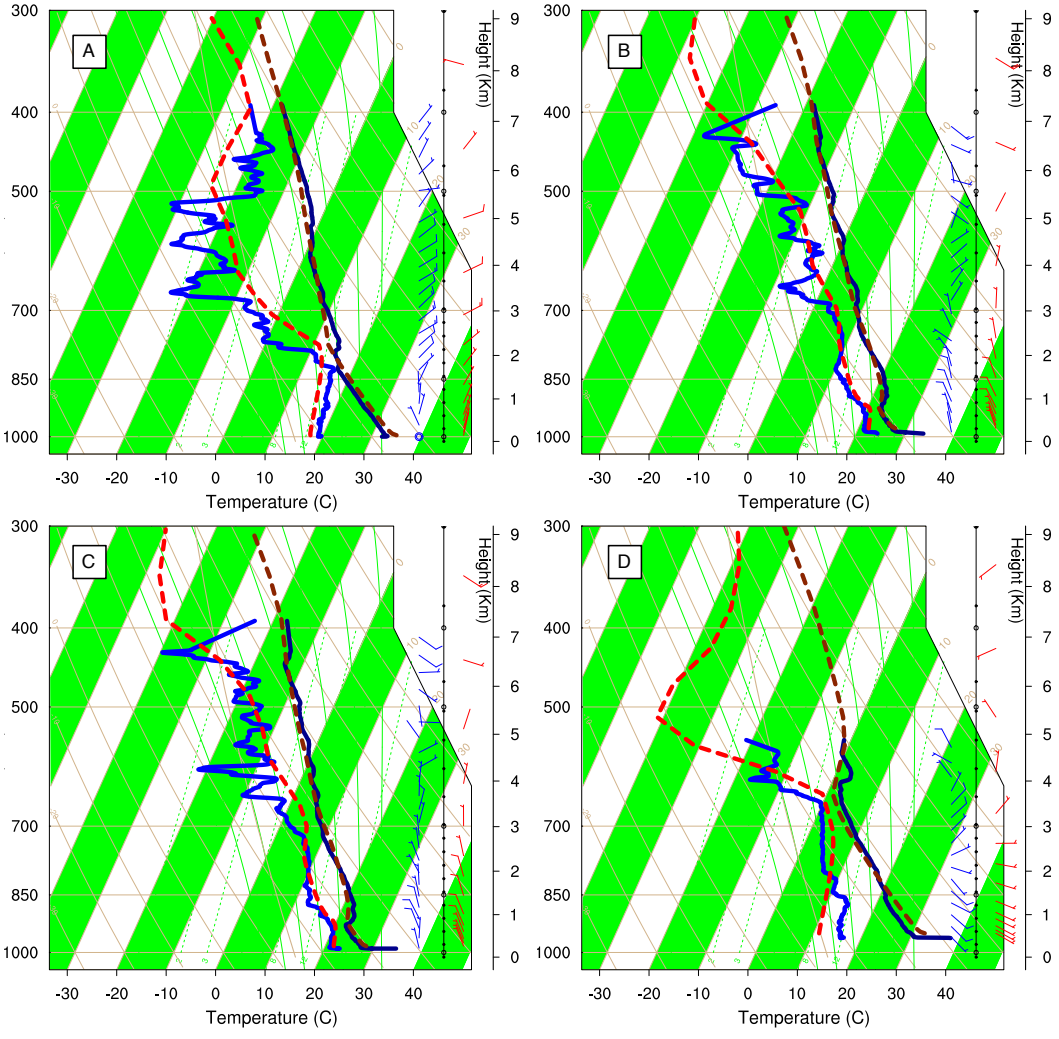

Figure 4. Skew-T plots comparing data from sondes dropped during SAMBBA flights with column data extracted from the WRF-Chem model at the time and place of the drop-sonde. Drop-sondes taken from **(a)** B731 (14 September, dropped at 16:02:28 UTC), **(b)** 734 (18 September, 12:46:52 UTC), **(c)** ~~B737~~ B734 (~~20~~ 18 September, ~~15~~12:~~23~~56:~~59~~53 UTC) and **(d)** B742 (27 September, 13:36:59 UTC). Red dashed lines from WRF-Chem model data, blue solid lines from drop-sonde. ~~Bight~~ Bright coloured lines on left show dewpoint (°C), dark coloured lines on right show temperature (°C). Barbs on right of plots show wind direction from drop-sonde (blue) and model (red).

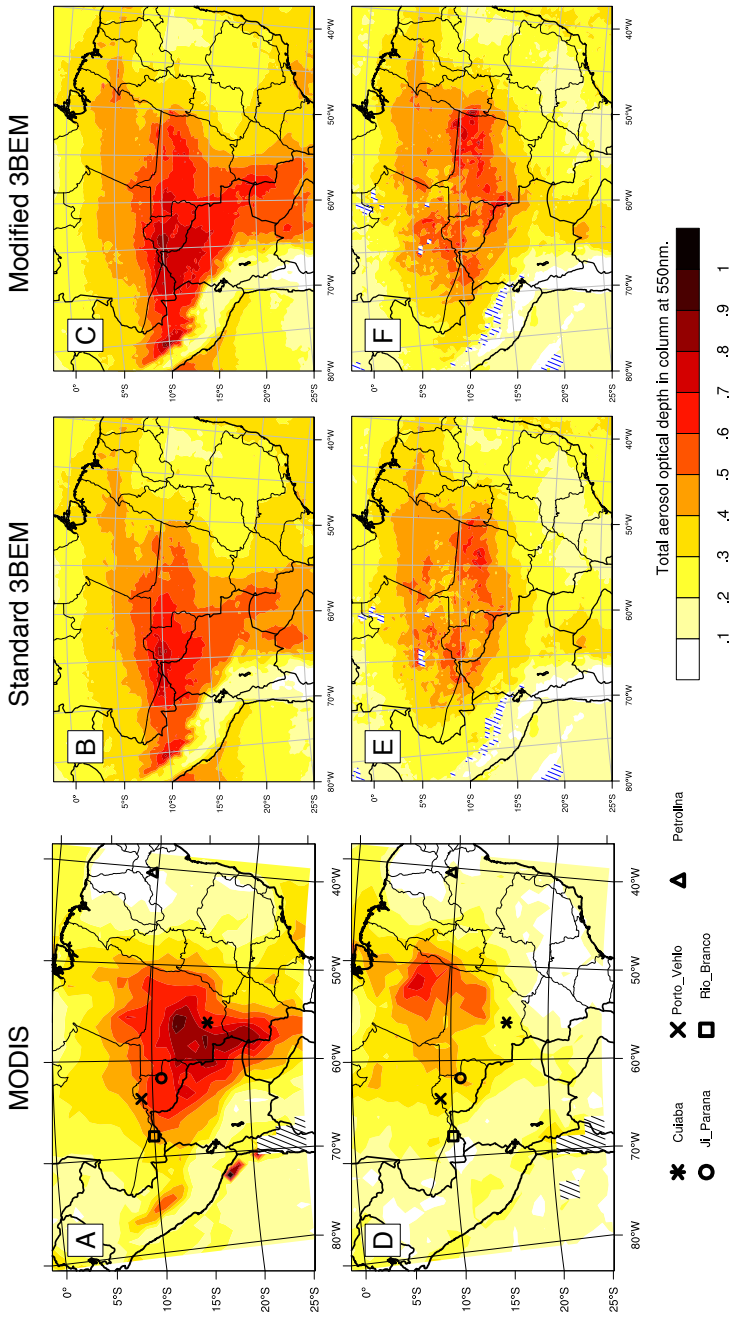

Figure 5. Horizontal ~~map~~ maps of column AOD at 550 nm, comparing the WRF-Chem model runs ~~agains~~ against MODIS measurements onboard the Aqua and Terra satellites. WRF-Chem data was extracted at times close to the overpass times of the Aqua and Terra satellites over South America. **(a**, **b** and **c)** for the first phase of the campaign (6–22 September 2012), **(d**, **e** and **f)** averaged over the second phase of the campaign (23–30 September). **(a** and **d)** combined ~~MODIS~~ Aqua and ~~TERRA~~ Terra satellite data~~.~~, **(b** and **e)** from model runs using standard 3BEM emissions~~.~~, **(c** and **f)** using ~~,~~ modified 3BEM emissions. The ~~location of~~ symbols in panels **(a** and **d)** signify the ~~sites~~ location of ~~the five operational~~ AERONET sites operational during the campaign period.

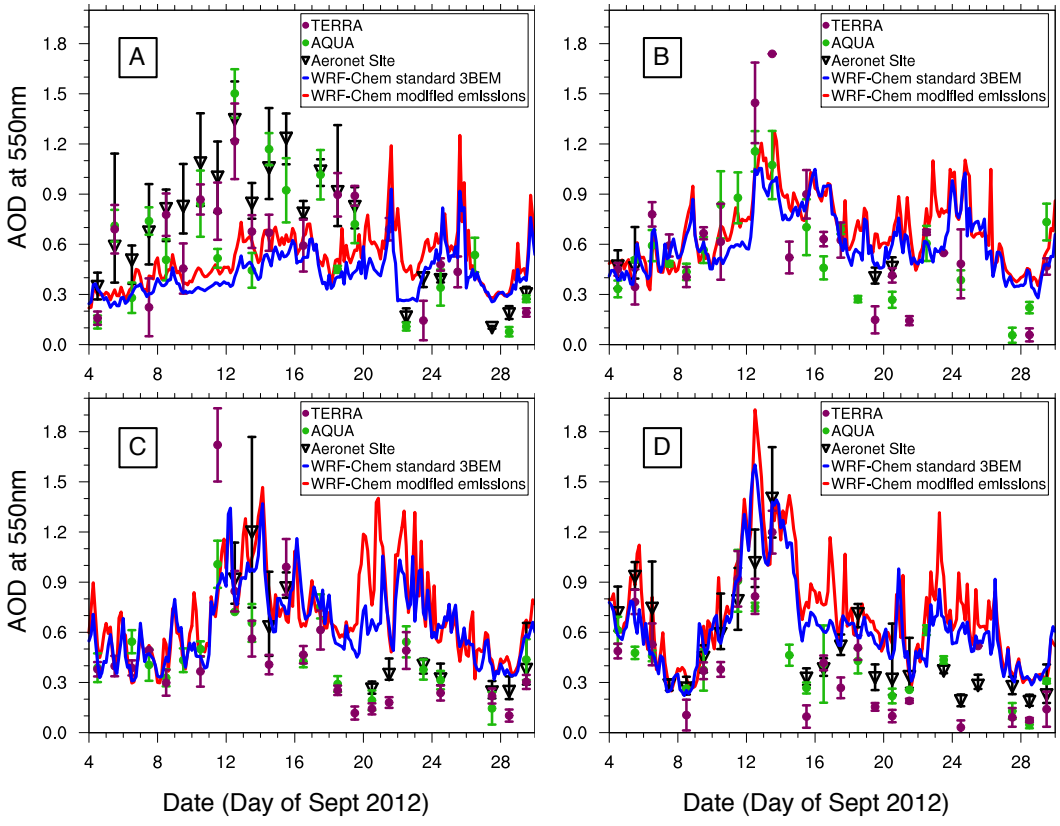

Figure 6. Timeseries of aerosol optical depth at 550 nm at four Aeronet sites between 4 September and 1 October 2012. ~~(a~~**(a)** ~~and b)~~ at Cuiaba, ~~(c and d)~~ **(b)** at Ji Parana, **(c)** at Porto Vehlo ~~, (c~~ and ~~f)~~ **(d)** at ~~Ji Parana and (g and h) at~~ Rio Branco. Blue triangles show Aeronet Site daily measurements, with bars indicating range in values over the day. Purple and green circles indicate measurements from overpasses of TERRA and AQUA satellites respectively, with bars indicating error range. ~~Red line shows~~ Blue lines show data from WRF-Chem model ~~, (a, c, d and f) from model run with traditional~~ simulations using standard 3BEM emissions~~, (b, d, f and h)~~ . Red lines show data from WRF-Chem model using the modified emissions.

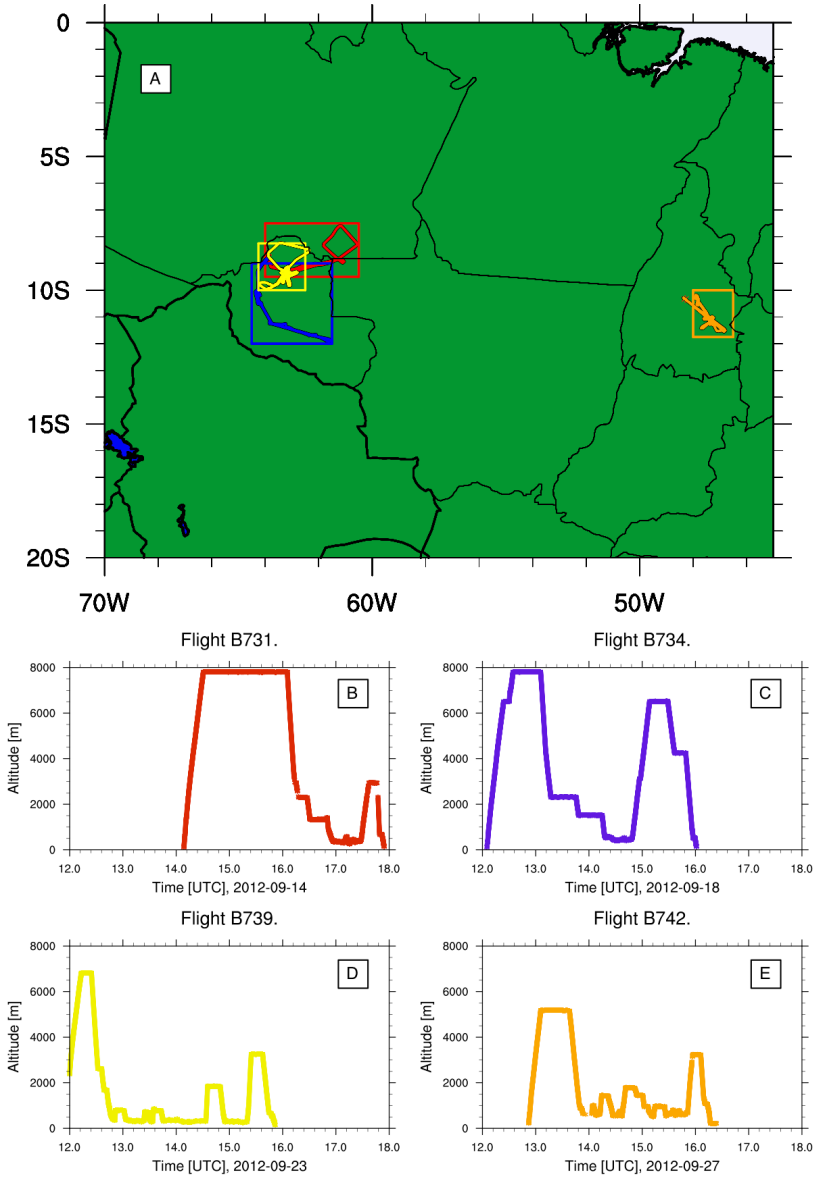

Figure 7. **(a)** Map of SAMBBA flight trajectories. Red: B731, 14 September 2012. Blue: B734, 18 September 2012. Yellow: B739, 23 September 2012. Orange: B742, 27 September 2012. Lines show path taken by flights, boxes show regions in model averaged over when comparing between model and flight data. **(b–e)**, altitude tracks of the four flights used for case-studies.

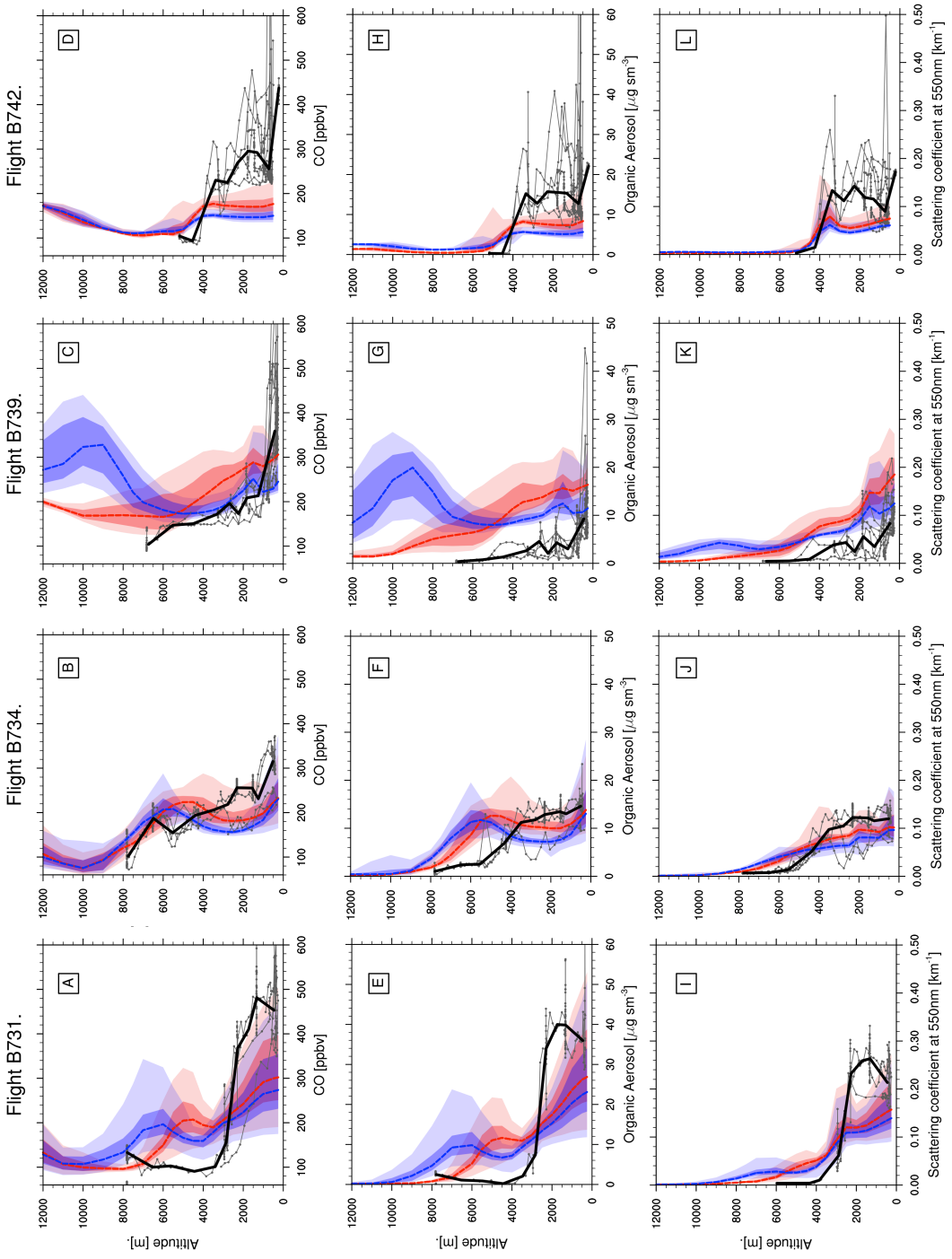

Figure 8. Vertical profiles of CO (ppbv), POM ($\mu g\,sm^{-3}$) and b_{scat} at 550 nm (km^{-1}). **(a**, **e** and **i)** from flight B731 (14 September 2012), **(b**, **f** and **j)** from flight B734 (18 September), **(c**, **g** and **k)** from flight B739 (23 September) and **(d**, **h** and **l)** from flight B742 (27 September). Red dashed lines show median from the modified emissions scenario, with strong red shaded region the interquartile range and the faded region the 5th–95th percentile range. Blue lines and shaded regions are for the standard 3BEM emissions scenario. Solid black line shows median line of profiles conducted by flights, fine grey lines flight measurements averaged over every 3 min.

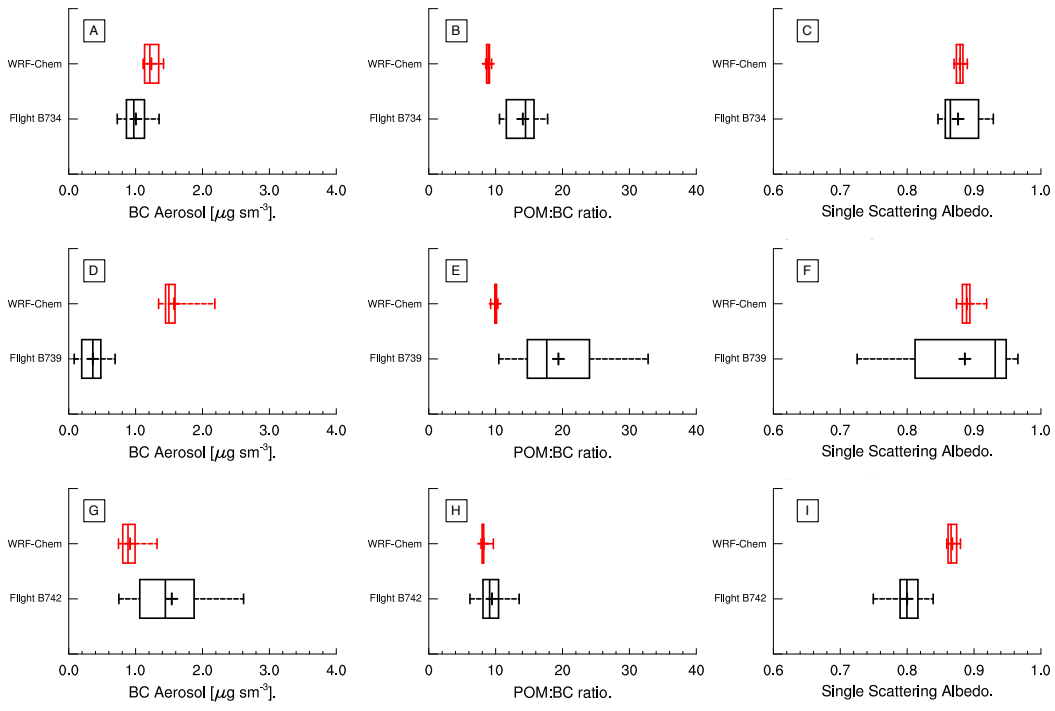

Figure 9. Box-whisker plots of black carbon (BC, $\mu g\,sm^{-3}$), particulate organic matter to black carbon ratio (POM : BC) and single scattering albedo (ω_0)~~, with~~. Box bounds ~~of box showing~~ show interquartile range, the end of dashed lines the 5th and 95th percentiles, and cross ~~over~~ indicates the mean. Showing spread of data from flights and extracted along flight path from modified emissions WRF-Chem run. Screened to only show data from straight-level runs below 3.25 km a.s.l.. Flight data averaged over ~~every~~ three ~~minutes~~ minute periods (approximately the time taken to travel across one 25 km grid cell). Panels **(a**, **d** and **g)** flight B734 (18 September), panels **(b**, **e** and **h)** flight B739 (23 September) and panels **(c**, **f** and **i)** flight B742 (27 September).

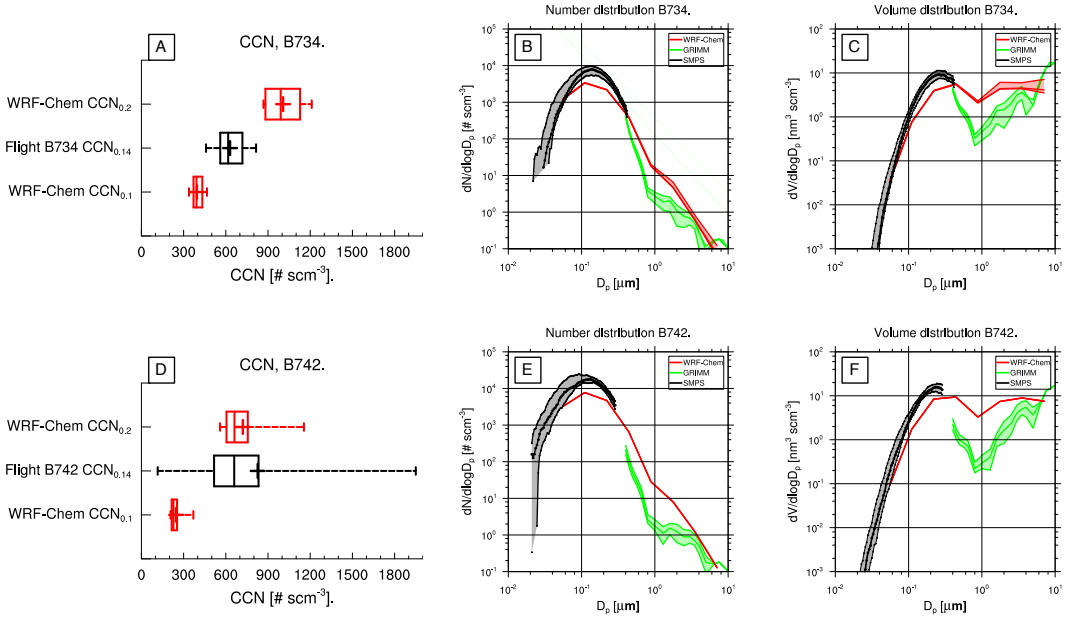

Figure 10. Plots of CCN concentration (scm^{-3}) and size distribution $\mathrm{d}N/\mathrm{d}\log_{10}(D_p)$ (scm^{-3}). Comparing flight data from flights B734 **(a**, **b** and ~~**b) c)**~~ and B742 ~~**(c (d**~~, **e** and ~~**d) f)** with model data from modified emissions run. Model data extracted along flight path and interpolated in vertical axis and in time. CCN plots show CCN concentration at approximately $0.14\,\%$ supersaturation ($\mathrm{CCN}_{0.14}$) from measurements, with CCN concentrations at $0.1\,\%$ and $0.2\,\%$ supersaturation ($\mathrm{CCN}_{0.1}$, $\mathrm{CCN}_{0.2}$) from model. ~~Size distribution shows red line for median WRF-Chem~~ Number and volume size distributions show data ~~over 8~~ from WRF-Chem modified emission scenario across the full 8-bin MOSAIC size ~~bins. Black line median from~~ range (red), the SMPS instrument ~~, green line median from~~ below 0.3 µm (black) and the GRIMM instrument above 0.3 µm (green). ~~Shaded~~ Central lines show median and shaded regions show interquartile range.