

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

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

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

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

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

Method description: Sect S2, Supplement

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

Results: Table S3, Supplement

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

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

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

Manuscript results, Sect 3.2:

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

Manuscript discussion, Sect 4.1:

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

### Minor Revisions and Technical Corrections

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

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

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

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

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

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

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

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

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

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

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

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

p. 8, l. 27. It seems surprising that CO at the surface decreases from W1 to W2 (from 340 to 220 ppbv) whereas CO<sub>2</sub> increases slightly (from 394 to 397 ppm). Does this suggest biogenic influence?

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

p. 9, l. 18. If this statement ('Significantly, the shift ...') is based on Fig. 9, should the end of the sentence actually read '... the relative abundances of rBC and sigma<sub>sp\_dry</sub> \*to\* CO'?

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

Significantly, the shift in meteorology between these two phases does not substantially impact the relative abundances of rBC,  $\sigma_{sp\_dry}$  and CO to each other.

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

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

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

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

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

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

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