Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-921-RC2, 2018

Atmospheric Chemistry and Physics Discussions

Interactive comment on "The vertical distribution of biomass burning pollution over tropical South America from aircraft in situ measurements during SAMBBA" *by* Eoghan Darbyshire et al.

Anonymous Referee #2

Received and published: 5 November 2018

Review of Darbyshire et al smoke vertical profiles in Brazil.

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

One point to raise is that there are two different important questions: 1) what is the state of the atmosphere? and 2) what causes it? The paper seems stronger on the

Printer-friendly version

Discussion paper

first point, and information in regard to the first question is important even if the second one cannot be answered definitively.

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

Issues to address include:

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

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

3) The potential effects of aerosol concentration and ambient temperature on gas-particle partitioning.

Clues to these issues might possibly come from e.g., the coating thickness to BC ratio, neph-scattering to BC ratio, neph-scatter:SP2-scatter?, and O:C ratio of any AMS spectra collected or SPMS data that may have been even occasionally collected.

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

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

6) Was any use made of GOES data?

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

board?

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

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

Comments by line number:

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

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

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

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

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

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

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

https://agupubs.onlinelibrary.wiley.com/doi/epdf/10.1029/2018GL078679

https://www.atmos-chem-phys.net/11/6787/2011/acp-11-6787-2011.html (See Table 1

on fire detection).

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

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

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

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

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

P6, L24: delete “c” in “cm-3”?

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

P7, L8&10: “lifted” > “lifting”

P7, L27: “Whist” > “Whilst”

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

P7, L29-32: Most of the fires are set by farmers, ranchers, or loggers, and they like to burn at low RH and wind speed to get good fuel consumption, but with reasonable ease of control/less chance of escape. An increase in either RH or wind speed means

less suitable conditions for initiating burning. However, also, increased cloud cover can reduce fire detection efficiency.

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

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

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

P8, L23: “surface” should be more precisely defined since we observed some instances of very high, reasonably widespread CO (∼5ppm) on the ground in the Amazon.

P8, L24: “unknown” or is e.g. “variable” more appropriate?

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

P8, L31: How is the “aerosol burden” calculated? E.g. is there an inferred mass? The aerosol emissions are usually higher at lower MCE in most studies, unless the authors have a good example in mind of the contrary? Seeing PM/CO (g/g/ or scat/ppm) larger at higher altitude could indicate PM evolution or that high MCE does not cause high plume rise? Other factors that influence plume rise, such as fuel consumption rate or atmospheric physics, might be more important?

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

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

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

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

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

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

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

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

P10, L9: “they exhibit”?

P10, L13: “eats” should be “east”? and L13-14: “above the lifting”?

P10, L22: In reality, it should be remembered that “plume injection altitude” is often not a fixed number, but an evolving mass-flux-vs-altitude, surface to plume top, distribution function (see Stocks et al (1996, JGR) SAFARI-92 paper).

P10, L25-34: A number of things besides MCE could influence “aerosol”/CO. For example, secondary organic or inorganic aerosol, temperature, the increase in MSC with aging, and fire intensity as noted in Reid et al., (1998) where more intense fires had much higher EF-PM similar to the finding for wild vs prescribed fires in Liu et al. (2017). Fires tend to be more intense later in the day when the mixing layer is also deeper. It’s also possible the authors have the relation between MCE and EFPM reversed as noted above. I don’t recall any of the cited works supporting the author’s claim that aerosol/CO increases with MCE, but if they do, it might help to point out the specific figure or table where that evidence occurs? The phrase “combustion completeness phase plume injection height” is awkward and at a minimum, perhaps missing a word

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

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

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

P11, L14-28: Good points.

P11, L29-33: I think the description of fires is potentially misleading and other driving factors for aerosol/CO noted above deserve mentioning here. Small fires in the tropics in homogeneous fuels often are mostly a steady mix of emissions entrained into a column by flaming. Residual smoldering could play a role, but explicitly accounting for post-convection, residual-smoldering emissions would require separate EF and separate fuel consumption, which is not trivial to estimate. Estimates are available in JGR papers by Bertschi et al 2003 and Christian et al 2007, specifically for Brazil. I think the key here is to characterize the structure and the uncertainty in the structure, with a list of possible causes (examples above) being nice, but perhaps presented without choosing a preferred option, unless additional evidence can be presented also.

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

P12, L1-12: I kind of glazed over trying to follow this, so I’ll just interject a detailed caution against assuming MCE and plume rise are positively correlated while freely admitting that they could be related at times.

Converting fuel carbon to CO2 releases more energy than converting it to CO, but

buoyancy is related to air temperature and the rate of fuel consumption or CO2 production. A fire burning with lower MCE could consume fuel at a higher rate in heavier fuels. Even for a fixed fuel loading, imagine a flame front at the beginning of a fire burning with an MCE near 1. As soon as it moves there is smoldering in its wake and MCE drops, but total CO2 is increasing for a relevant-sized area because more fuel is burning. Air temperature above the fire peaks when CO2 peaks, not MCE. That is what has been seen in most of our lab experiments or field work with towers or low flying helicopters. E.g. in Akagi et al 2012 an 81 ha fire had similar MCE at different heights in the convection column and the plume rise was dependent more or less on the instantaneous area burning. If you watch tropical fires, plume rise often peaks when flame fronts started on opposing edges converge, and then they annihilate.

Secondly, even if the plume has more thermal energy, it may not rise higher ultimately. Plume rise depends on atmospheric stability, entrainment, the vertical profile of the winds, temperature, moisture (condensation can release energy), etc. The authors are in a good position to advise on the typical thermodynamics!

Thirdly, flaming vs smoldering may impact the plume rise, but to check further maybe try some other F/S indicators like BC/CO that might work better or independently.

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

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

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

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

Fig 3. There are two BB plumes identified on the rBC trace. There are no BB plumes identified on the CO trace, and only one each on CO2 and Bscat. It seems like there

should be two on all traces if they really are BB. Also, I find it hard to locate/digest the structure and plume features that are explained in the upper right boxes.

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

Fig 5. Maybe label FC as “FC/100” also in Fig 6? Interesting that the AOD max is downwind of the detected fire max. This could be transport, but also different detection efficiency, higher PM emissions from more smoldering in forest fires. AODs up to 4 that are mentioned in the text relate to this figure how?

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

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

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

Fig 9. Needs units. More?

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

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