

Our responses to Jeffrey's Reid's comments are detailed below. Reviewer's comments are in *italics* and our responses in standard font.

General comments

1.

The authors may want to review their history a bit for their introduction, as this I think is important in the interpretation of their data. I found quite a few factual errors listed, and this leads to some misinterpretation of their data. Prior to SCAR-C (1994) and then SCAR-B (1995), the only mechanism of particle growth in biomass burning plumes thoroughly considered was coagulation (in the biggest fires this is still likely to be true).. A very good example of how things were thought to evolve is in Radke's 1995 paper "Effects of aging on the smoke from a large forest fire" in Atmospheric research, [http://dx.doi.org/10.1016/0169-8095\(95\)00003-A](http://dx.doi.org/10.1016/0169-8095(95)00003-A) . This is a very good and relevant read for you, even though I am not so sure he had lagrangian pairs in there. It was my 1998 paper (Reid et al., 1998 as you reference page 6448 line 15) that was the first to point out that the dominant aspect of growth was not coagulation, but condensation. Even this was a bit of a fight with my advisors, because SOA yields were thought to be on the order of a percent or two, and there were certainly not enough VOCs out there. So at the time, I pushed for condensation of long chain hydrocarbons, based on the fact that we found particle emissions factors a factor of two higher at the top of a smoke plume compared to the Darrel Ward towers at the bottom. Also, based on Vanderlei Martins SCAR-C and my SCAR-B electron micrographs, we could see that particles were getting coated in organic goo in an hour or so. At the time however, we did not recognize the important role of oxygenated hydrocarbons, which I think is the preferred source (although I have not entirely given up). A key point here, is that SOA and or condensation processes happen very rapidly, like on the order of hours after emission. At the same time as this was going on Cathy Lioussé was publishing her work on fire monitoring in Africa-see Lioussé "Aging of savannah biomass burning aerosol: Consequences on their optical properties" J. Atmos. Chem. <http://link.springer.com/article/10.1007%2F978-3-70-007081-7> . This process is probably on the same order or a bit longer than growth. But probably no more than a day. Since then the community has gone back and forth on what is the significance of the condensation/Secondary Organic Aerosol (SOA) versus evaporation. Personally I think it is on average what I said in 1998, where from the "top of the smoke column" to a day downwind is on the order of 20-40% mass growth with a substantial fraction of this is being inorganic (and this requiring cloud processing). Thus, while this is substantial in things like emission factors or ultimate radiative impacts of smoke, it is only about a ~10% increase in particle size. Or, going from a VMD of 0.32-0.355 μm . Some people say this is too much, some say this is too little. Nevertheless, I think it is a good baseline from which you do an uncertainty analysis.

Reply:

Thank you for this in-depth and very insightful comment. We have perhaps misinterpreted the time scales of particle growth processes and their relative importance towards the total size growth. As a result we have initially attributed the apparent increase in particle size both to coagulation and condensation giving too much weight to condensation. Admittedly, there is not much in our data support or reject condensation and we relied on other studies making the inferences. Our misinterpretation was influenced by studies indicating substantial organic coatings in well aged plumes (for example Dahlkötter et al., 2013). Also it appears that we have misread the reviewers work from 1998 in Amazon (Reid et al., 1998), which states:

" In regional hazes, over periods of 1 to 4 days, particle coagulation and

condensation probably contribute about equally to particle growth. After 3 days, most of the condensation and gas-to-particle conversion has likely taken place, in which case coagulation would be the only significant particle growth mechanism."

We have revised the introduction paragraph discussing ageing effects, including suggested literature and clarifying the particle growth processes and time scales at which they occur:

"Most of the changes occur within minutes up to a few hours after emission. Above the flaming zone in cooling plumes particles grow rapidly in size and mass by condensation and coagulation. During the first few hours particle distribution volume median radius has been reported to increase by up to ~ 60 % (Hobbs et al., 1996; Abel et al., 2003; Calvo et al., 2010; Akagi et al., 2012). Hobbs et al. (1996) measured a growth in volume median radius from 0.125 to 0.19 μ m in two hours for a large and intense prescribed burn in North America. Near source condensation of low pressure vapour organics and secondary production of inorganic and organic particulate matter are thought to increase smoke particle size by up to ~ 10 % (Reid et al., 1998). The rate of coagulation is approximately proportional to the square of particle concentration (Lee and Chen, 1989) and is, therefore, highest near to the source. In highly concentrated plumes, however, coagulation can be important on the time scales of days. Particles continue to grow on these time scales, but at a much lower rates (Radke et al., 1995; Reid et al., 1998; Capes et al., 2008)."

The interpretations of the results was modified stating that the primary mechanism for long term particle growth in large dense plumes is coagulation.

2.

The next question then is the timescale coagulation. Here, coagulation because relaly impronant for high concentrations for long periods of time. Indeed, in my dissertation 20 years ago I downplayed coagulation's role except for in the large continental super plumes, which in fact this paper is looking at are looking at. Now the real trick is at what time scale all of these things happen. As I mentioned above, I think a big chunk of the secondary particle action is oin the 2 hours. Regardless of your persuasion on condensation and SOA production, I have never seen anything along the lines of rapid mass growth longer than half a day. The problem is that AERONET cannot perform a retrieval under these circumstances. Even if one were lucky and had a site right next to the source, the sky would not be uniform. Thus, this system is likely suitable for evaluating the evolution of moderately to well-aged smoke, not from source to well-aged. This would be a coagulation dominated region. Of course, the bulk of the community and I could be (and are frequently) wrong about such things

Replay:

We have refined our interpretation of the inferred particle growth reflecting the above clarifications. Notably, there was a paragraph discussing the limitations of the AERONET data and our method characterising fresh smoke. To make this aspect more clear we have extended it stating:

"The AERONET records typically do not include observations of truly fresh smoke within seconds or minutes after the emission. Consequently, our results are for young to well aged smoke, which is already transformed by the rapid initial growth and has generally large particles."

To avoid any further confusion regarding ageing time scales we have replaced the

term "fresh smoke" with "young smoke" throughout the manuscript.

3.

But from a point of view of this system, it should be clearly put as a likely aspect of the biomass system that is being analyzed, the inherent sampling bias that occurs, and how then such data should be interpreted by the community. So considering the above information, then interpretation of the data because a bit easier. First, from a sampling point of view, the plume must be big enough to allow for two points to be compared. This can only be done then for large boreal and mid-latitude fires. If the fire is to be detected 144 hours downwind as the dominant aerosol specie, the sampling bias is then extreme. These have to be truly massive and hence dense, and thus coagulation will be enhanced.

Reply

A discussion of sampling bias has been added to the results section, clarifying that the results are for large plumes:

"The method employed was limited to tracking of highly concentrated free tropospheric plumes emitted from fires larger than 100km² and the results are representative of such events. The sampling bias is particularly severe for very old attributions as only very dense continental superplumes can be observed after several days of ageing."

To address the sampling bias for older observations in the analysis, we have introduced a comparison of ageing effects for extremely optically thick (white points in fig. 6a) and less thick plumes, identified by the highest satellite retrieved value along the trajectory:

"The estimate is only an indication of initial plume concentrations because of limitations retrieving AOT over optically thick plumes and large uncertainties associated with high AOT retrievals. However, the highest AOT values are typically found within hours from the source, and therefore are better indicators of the initial plume concentrations than the downwind AERONET AOT retrievals. Pooled plume concentration estimates seen in Fig. 5d exhibit bimodality. The bulk of maximum AOT values are centred around 1.5, but approximately a quarter of the plumes indicate extreme optical thickness with maximum AOT values close to or at MODIS saturation value of 5.0."

4.

I am not sure I would interpret Figure 6 as cleanly as presented. If you look at the combined work of Turko from the 1990's, condensation will narrow a volume distribution, whereas coagulation will keep it study. But what we find in figure 6 d is that really for any given age the standard deviation increases with VMD. Thus, this is likely a nature of the "source" rather than aging-although I am basing this interpretation on a very small scatter plot.

Reply

We disagree with this interpretation of the figure 6 d. Admittedly, it may be not clear from the given scatter plot, but what we see is that the approximately linear relationship between VMR and spread (R-squared = 0.45, p = 0.000) for relatively young smoke observations (< 1 days old) is not evident for older plumes (R-squared = 0.005, p = 0.2). The change is mostly occurring along horizontal axis (VMR). We have changed the figure (6b in the updated manuscript) showing young and well-aged (> 72h) plumes as two populations:

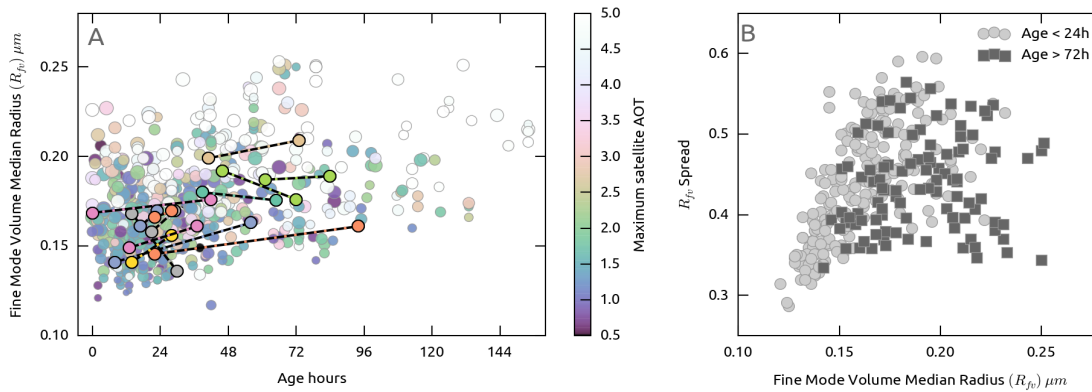


Figure 6. (a) Fine mode volume median radius and estimated age. Points joined by lines show identified paired observations. (b) Fine mode volume radius against the spread of fine mode volume radius for young and well aged plumes.

5.

Second then is the correlation in Figure 6 b, whereas aged plumes with higher AOT appear to have higher angstrom exponents pass 96 hours. Again, this may be a sampling issue. Less than 96 hrs, I am not sure there is any correlation at all except for very high AOTs (AOT>1). This is probably mostly real, although to me it looks more like two populations than something you would want to fit with a regression.

Reply

The relationship between age and absorption angstrom exponent shown in figure 6b is indeed weak. It has been removed from the analysis. Notably, responding to this and other comments, we have changed the way our results are presented. Instead using linear regression to infer the ageing effects, we compare plumes split into broader age categories. We believe that such method is more suitable for the data and represents the results more realistically, without implying linearity.

6.

I suggest the authors have a look at Edward Hyer's recent work, that lays out that source attribution is not so east from space, based on a combined error in land cover, navigational error and temporal sampling (e.g., <http://onlinelibrary.wiley.com/doi/10.1029/2008GL036767/abstract>;)

Reply

Thank you for pointing to this work. The source estimates presented have errors at several levels. As pointed out, the uncertainty of the MODIS land cover type products, large within fire object and within pixel variability in land cover type, and unquantified uncertainty in the back trajectory analysis and source age attribution method. We have expanded the paragraph discussing the uncertainties associated with the source attribution:

"The source estimates presented here have uncertainties at several levels. Hyer and Reid (2009) found that MODIS land cover type products was only accurate in ~88% of the cases analysed. In addition, only the dominant land cover type for a given fire is considered here ignoring varying proportions of included grids attributed to different vegetation and intra-grid mixing. Finally, unquantified

uncertainty in the back trajectory analysis and source age attribution presented here."

7.

I would also be very careful with the interpretation of PWV and smoke age. This has been seen many times in the past (I would add Remers work over the Amazon which kicked this off) and is usually attributable to confounding. Indeed, is the smoke getting untrapped into moister airmasses, or is the dry smoke layer aloft transporting over a moister airmass? Besides, PWV often has no bearing on RH which is what drives hygroscopicity. Looking at Figure 6C I am not sure there is so much to hang your hat on.

Reply

Admittedly, whilst the relationship between PWV and age is significant, it is a weak one. We have removed it from the results section, focusing on aspects that are more clear and easier to interpret.

8.

Finally I would just ask that a few details be placed in the real nature of remote sensing. Little pieces of information are misleading. For example, when you say the MODIS aerosol product is 10x10 km, that is at nadir. It is on average twice that given the scan angle.

Reply

This has been clarified updating the data and methods section:

"Both data products provide interpolated AOT at 550 nm with 10 km × 10 km pixel size at nadir, which is doubled at the edge of swath for MODIS and increase only negligibly for AATSR."

This aspect, however, does not influence the results significantly, as the satellite data is used in the analysis only qualitatively.

9.

While they note that the errors in the MODIS product are skewed towards clean conditions and that for fires errors may be extreme, they might also note that MODIS cannot do smoke retrievals near a fire in the first place, except on the edges of a plume

Reply

We have clarified the uncertainties associated with satellite AOT retrievals over optically thick plumes:

"...the algorithms do not estimate AOT over opaque plumes near the source and often reject bright dense smoke as cloud or bright surface (Livingston et al., 2014)."

This caveat affects this study to a lesser extent. Continental scale plumes are dispersed comparably fast and can be observed for days with enough AOT retrievals to identify and track the plume in many cases.