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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Smoke aerosol properties and ageing effects for Northern temperate and boreal regions derived from AERONET source and age attribution" *by* T. Nikonovas et al.

Anonymous Referee #1

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Summary:

This paper uses AERONET inversions sampling northern hemisphere biomass burning aerosols together with HYSPLIT back-trajectories and satellite AOT/fire count data to attempt to quantify ageing effects on smoke aerosol optical and microphysical properties. The data are also stratified into seven source land cover types and analysed. The topic is important and relevant to ACP, and the quality of English is good.

I would like to note I am in the radiative transfer/ remote sensing community, so my

comments focus on these aspects rather than the biological/chemical side of things.

AERONET inversions have been used widely for radiative transfer applications, for example for forcing calculations or to build optical models needed as constraints for satellite AOT retrieval algorithms, and there are some advantages and disadvantages of AERONET as a data source for this type of analysis. The main new aspect of this work, in my view, is the use of HYSPLIT and satellite data to track individual events and quantify potential ageing effects. This has been looked at for a few individual case studies (e.g. cited in the paper) but not to my knowledge on such a large scale. The results seem to indicate an increase in fine-mode particle size as plumes age, covarying with an increase in columnar water vapour, which affects the spectral/directional dependence of extinction somewhat but less so the SSA.

The basic details of the method (in terms of tracking trajectories) seems sound to me. However I think that a better discussion of the uncertainties on the AERONET data and the analysis itself is required. Many of the uncertainties here are not the authors' fault: there are uncertainties associated with the AERONET inversions, and the small data counts mean that regression relationships presented are limited in what they can say. For example, about 73% (going by Figure 1) of the inversions sampled smoke 0 or 1 days old, and only about 6% sample smoke 4 or more days old. In this sense the prominence of 'ageing effects' in the paper title is perhaps getting our hopes up too much. Indeed, perhaps these limitations are the reasons why a study on this scale has not been performed before. Perhaps it would be better to look at the events contributing to these 6% in more detail, to separate out near-source variability and ageing effects better.

Because of this, I favour revisions as discussed below. I would be willing to review a revised version of this manuscript.

Specific comments:

P6451L15-16: Strictly, the statement that level 2 inversions contain only retrievals for

AOT(440) of 0.4 or higher is wrong. Cases of lower AOT are still included in the data product as provided on the AERONET website, it is just that information related to e.g. SSA is removed because this is thought to be unreliable. The size distributions are still there. One possible way to increase the data volume of aged plumes in the analysis would be to look at the inversions below this AOT=0.4 limit. It would not help the SSA analysis, but could help the analysis about particle size, Angstrom exponent, precipitable water, and AOT changes.

P6458, L6: Here and elsewhere the authors state that the AERONET SSA uncertainty is 0.02. I am puzzled where this number comes from (no references are provided by the authors here). The 'canonical' number (Table 4 of Dubovik et al., JGR 2000) is 0.03 for biomass burning aerosols, for cases where AOT(440) is 0.5 or higher. It's also not apparent how much of that is systematic, and how much is random, error. I realise that this will probably not affect the interpretation of results since a main conclusion appears to be that the results for most surface types are not significantly different from SSA=0.95, but nonetheless I think this should either be corrected, or a source for the SSA uncertainty being 0.02 rather than 0.03 provided here.

Figure 6: A lot of the discussion on ageing effects in the text relates to this figure. Confidence intervals (CI) are drawn based on linear regression fits to the data. However it is clear that the ageing effects are small compared to the scatter in the data. So these relationships are explaining only a fraction of the variability. In this sense I think discussions of the CI could mislead a casual reader – these are the CI on the gradient, but could be mistaken to imply something about the spread of the underlying data as well. For this reason I think it would be useful to also provide the coefficient of determination for these plots (and in the text).

Related to the CI, how exactly where these calculated? This is related to the assumptions about the uncertainty on the y-axis data going in. Were these taken to be the AERONET uncertainties, or weighted equally, or what? Were data points assumed to be uncorrelated? Were uncertainties on regression coefficients scaled by the reduced

chi-square value (which is the equivalent to assuming that the regression model is 'correct') or not? This information should be provided in the paper. I'm also curious about the two-sided p threshold of 0.01 being used to denote significance when CI are presented as 95% bounds (which I would think corresponds to p values of 0.025, or am I wrong)? Why the inconsistency here?

I would also point out that statistical significance does not necessarily correspond to scientific significance (for example, if an effect can be discerned, but it is so small as to be negligible for practical purposes). Equally a number may be large but have a huge error bar, in which case all you can say is that you don't know what's going on.

As hinted earlier, one way to cut through some of this scatter would be to look at some case studies in more detail. Figure 1 says there are 6 cases with an estimated smoke age of 6 days. I am not completely clear if this means that there are 6 cases where the same plume was observed more than once during these 6 days, or just that there are 6 cases where a plume that was 6 days old was observed. If the former (or if some of these events were sampled more than once), analogous plots to Figure 6 could be created for each of these case studies (rather than just looking at all points at once). Then we could see whether the noise in these plots is still present or not. That will say something about whether ageing effects are for example stronger or weaker than the AERONET inversion uncertainties (which is not really discussed for e.g. size distribution). I also worry from Figure 6 that the data are not suitable for linear regression analyses (due to non-uniformity of sampling along the age axis, and non-Gaussian behaviour of departures from the best-fit line); reducing to a subset of data corresponding to these few case studies would help with this issue.

Figure 7: For panels (e-g), it would be good to show the 'all classes combined' data in a heavier font/line, as at the moment it is hard to see among all the other lines. I also think that the data from this plot should be placed in a table, in addition to the Figure.

General throughout: 'angstrom' is not typeset correctly. In LaTeX, I think this should be

\AA{}ngstr\'{o}m.

Conclusion: Here (and in the introduction) the authors mention the need for better aerosol optical models for satellite retrieval algorithms. This is undoubtedly true. However as a practical matter, it is not clear how this new information could be used in (for example) a satellite AOT retrieval algorithm, because the instantaneous satellite snapshot will not 'know' how old the smoke is or where it came from, and it's not practical (might not even be possible) to use ancillary data to successfully identify smoke age and/or origin on a case-by-case basis for a global algorithm. Even if it were possible, the authors do not present evidence that including these ageing effects would make a significant difference to these retrievals (for example there are no radiative transfer simulations provided in this study), especially because (as mentioned) the ageing effects appear to be smaller than the scatter in the data (Figure 6). Sayer et al (ACP 2014) did some analysis in this regard looking at intra/inter-site variability in smoke aerosol models, and AOT errors resulting from the assumption of the wrong aerosol model, although this was mainly driven by SSA considerations (from virtually nonabsorbing through to strongly absorbing) so those simulations are not directly transferable to the case here (where there is little SSA variability and the main ageing effect, or difference between sources, is a change in fine mode particle size). On the modelling side of things, similarly, do these small ageing changes or differences in properties between different smoke sources result in meaningfully different calculations of e.g. shortwave flux or other relevant quantities? I realise that doing this in detail would probably be out of the scope of the manuscript, but found the authors' comments here to be a bit careless given the lack of substantiation, and the large error bars on the results of the analysis. The authors say that these new results can help but don't give a specific or quantitative look at how. I would prefer to see a deeper discussion here, or else remove the statements from the manuscript.

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