Our responses to anonymous reviewer's #1 comments are detailed below. Reviewer's comments are in *italics* and our responses in standard font.

General comment:

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.

Reply

Thank you for a very valid and constructive review. We have attempted to address all of the issues raised in this, and in following specific comments.
A discussion of uncertainties associated with individual AERONET retrievals has been added to the data section:

"AERONET uncertainty for individual SSA retrievals in these cases is approximately 0.03 (Dubovik et al., 2000). Particle fine mode volume median radius has an uncertainty of 0.01μ m , the spread of the fine mode particle distribution – 0.06. Estimated uncertainty in asymmetry parameter ranges from 0.015 at AOT(440) to 0.04 at AOT(1020) (Dubovik et al., 2000; Sayer et al., 2014)."

To account for uneven sampling along the time axis we have changed the way the results are presented. Instead of using linear regression coefficients, the ageing effects are discussed comparing differences in distributions of plumes grouped into broader age categories. The small number of very old estimates indeed limits what we can say about differences beyond the first 4 days of ageing. Therefore plumes older than 72 hours (more than 3 days) were pooled into one category and compared to smoke aged for less than 1, 2 and 3 days. We believe that such analysis better suits the data and represents ageing effects more realistically avoiding the pitfalls of the linear regression model.

Specific comments:

1.

P6451 L15-16: 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.

Reply

This was a misleading statement indeed. The authors were aware that the level

2 inversions for retrievals at AOT(440) lower than 0.4 contain information on many parameters except SSA and complex refractive index. The AOT(440)=>0.4 data selection threshold was used throughout because analysis of ageing effects on SSA was one of the main interests of the study when conceived. We do agree that extending the analysis with AOT(440)<0.4 cases could be beneficial in reducing the uncertainty in our estimates. However, exploration of these observations indicated that source attribution was much more difficult for the optically thin plumes. We were gaining a small number of identifications at disproportionally increasing processing costs. Importantly, looking at the additional points it was obvious that the scatter was still there and we felt that further processing would not have a significant effect on the main conclusions of the study.

The statement has now been removed. Instead it is stated that "AERONET level 2 inversions contain SSA retrievals only at AOT(440) levels of 0.4 or greater." Section 2.1.1 (Data selection) was updated stating that only retrievals at AOT(440))=>0.4 were used in the analysis.

2.

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.

Reply

We apologize for this human mistake. The number has been corrected where applicable, providing the reference at the first occurrence.

3.

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).

Reply

As stated above, we have since abandoned the linear regression analysis, hence this does not apply any more.

4.

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.

Reply

Confidence intervals for any quantity discussed (and previously for regression coefficients) were derived using bias-corrected non-parametric bootstrap method (Efron, 1993). During the resampling y-axis error was modeled as normally distributed AERONET uncertainty given as 1 standard deviation. We have now clarified this in the methods section.

5.

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.

Reply

We have reviewed the manuscript and removed the relationships (AAE, precipitable water), which are not only weak, but difficult to explain as well. The remaining ones (fine mode median radius, AE, asymmetry parameter, SSA) are interesting 1) either way if differences comparing young and well aged plumes are significant or not, and 2) agree with existing estimates. We do not present changes in ageing plumes for all land cover types, as indeed the uncertainties were large because of small number of plumes attributed to most of the classes. We have integrated AERONET uncertainties in our confidence interval calculations, and provide them for all of the estimates.

6.

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?

Reply

The mismatch has been corrected. 95% confidence intervals are given throughout.

7.

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)

Reply

We perhaps didn't make it clear enough, but we initially did not include

discussion of lagrangian pairs -- observations of the same plume at two or more AERONET stations. However, there were a few cases of identified paired

observations of the same smoke, the discussion of which has been now added:

“The trajectory analysis indicates that in several cases the same plume was transported over more than one AERONET station allowing to infer changes in plume properties between the two observations. Unfortunately, only 13 of such events were identified preventing a more robust analysis (Fig. 6a). In 10 out of the 13 cases older particles are larger, while hree of the pairs suggest a decrease in R fv between the observations. The median R fv change rate is 0.0075 (-0.001–0.03) μ m per day. The estimate agrees well with the growth rate suggested by differences in particle distributions between young and aged plumes. However, it has large confidence intervals due to low number of paired observations and uncertainty in individual AERONET retrievals.”

We did not present these cases initially, and do not want to give it too much weight now, because of their small number and several problems associated with such identifications. For very large plumes it is somewhat easier to determine source and approximate age than it is to identify paired AERONET retrievals of the same air parcel. The trajectory uncertainty analysis indicates that the trajectory window can grow to hundreds or thousands of kilometres in a matter of hours or days, and thus it is not possible to say that the two observations along the trajectory are of the "same" smoke with any degree of certainty. Allowing bigger trajectory windows yields more identifications, increasing false positives as well. We avoided having to inspect individual cases and selectively pick the good looking ones. As a result there were only a handful of such identifications. Importantly, AERONET uncertainty on individual retrievals is more important comparing a pair of observations (or a few pairs), and results in huge confidence intervals on the estimate. For the 13 paired observations that we had, the median slope in volume median radius is reasonable and agrees well with inferred particle growth. However, when AERONET uncertainty is factored in, the estimate is not significant. Comparing larger groups of observations helps to deal with this, and we are able to say with reasonable confidence that on average, older plumes have larger particles. We believe that such interpretation of the data is more appropriate, and have revised the manuscript accordingly.

8.

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.

Reply

This is obsolete as the linear regression analysis has been abandoned.

9.

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.

Reply

The figure 7 has been replaced by another as we no longer present change rates

derived from linear regression analysis. The new figure should be clearer as the number of parameters and classes we present has been reduced.

10.

General throughout: ‘angstrom’ is not typeset correctly. In LaTeX, I think this should be \AA{}ngstr\’{o}m.

Reply

Thank you for picking this up. The word has been changed throughout.

11.

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.

Reply

The statements have been removed.

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 qyite 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 . 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 Liousse was publishing her work on fire monitoring in Africa-see Liousse "Aging of savannah biomass burning aerosol: Consequences on their optical properties" J. Atmos. Chem. http://link.springer.com/article/10.1007%2FBF00708178 . This process is plrobalby 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 side. Or, going from a VMD of 0.32-0.355 um. 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 to 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 Dahlkotter 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 improtnant 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 12initial 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 form 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:

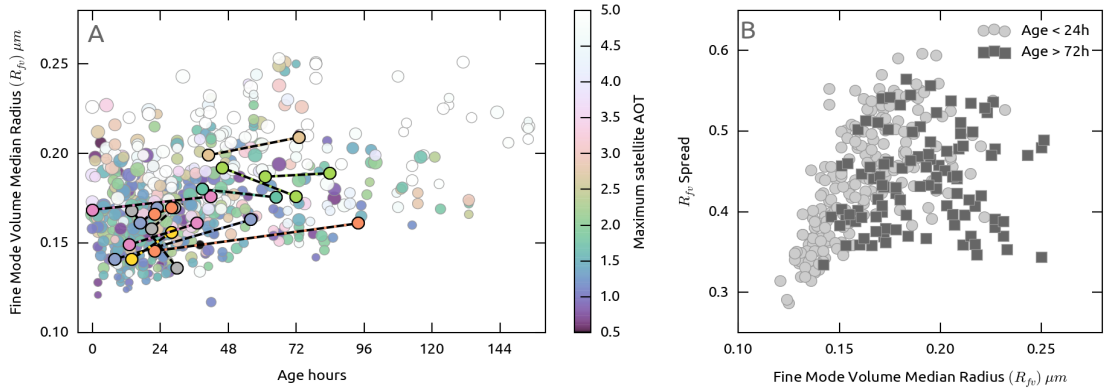

Figure 6. (a) Fine mode volume median radius and estimated age. Points joined by lines show identified paired observations. (b) Fine mode volume radius against the spread of fine mode volume radius for young and well aged plumes.

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 untrained 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.

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Smoke aerosol properties and ageing effects for Northern temperate and boreal regions derived from AERONET source and age attribution

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Abstract

Particulate emissions from wildfires impact human health and have a large but uncertain effect on climate. Modelling schemes depend on information about emission factors, emitted particle microphysical and optical properties and ageing effects, while satellite retrieval algorithms make use of characteristic aerosol models to improve retrieval. Ground based remote sensing provides detailed aerosol characterisation, but does not contain information on source. Here, a method is presented to estimate plume origin land cover type and age for AERONET aerosol observations, employing trajectory modelling using the HYSPLIT model, and satellite active fire and aerosol optical thickness (AOT) observations from MODIS and AATSR. It is applied to AERONET stations located in or near Northern temperate and boreal forests, for the period 2002–2013. The results from 629 fire attributions indicate significant differences in size distributions and particle optical properties between different land cover types and plume age. Smallest fine mode median radius (R_{fv}) are attributed to plumes from cropland – natural vegetation mosaic (0.143 µm) and grasslands (~~0.147~~0.157 µm) fires. ~~Evergreen~~ North American evergreen needleleaf forest emissions show a significantly smaller ~~fine mode median radius~~ R_{fv} (0.164 µm) than plumes from ~~woody savannas (0.184~~Eurasian mixed forests (0.193 µm) and ~~mixed forest (0.193~~plumes attributed to the land cover types with sparse tree cover – open shrubland (0.185 µm) and woody savannas (0.184 µm)~~fires. Smoke plumes are~~ . The differences in size distributions are related to inferred variability in plume concentrations between the land cover types. Smoke is predominantly scattering for all of the classes with median single scattering albedo at 440 nm (SSA(440)) values close to 0.95 except the cropland emissions which have a SSA(440) value of 0.9. ~~Overall fine mode volume median radius increase rate is 0.0095per day for the first~~ Plumes aged for 4 days ~~of ageing and 0.0084~~or older have median R_{fv} larger by ~0.02 µm ~~per day for seven days of ageing. Changes~~ compared to young smoke. Differences in size were consistent with a decrease in ~~Angstrom~~ Ångström Exponent and increase in Asymmetry parameter. ~~No significant changes~~ Only insignificant increase in SSA(λ) with ageing ~~were found. These estimates have implications for improved modelling~~

~~of aerosol radiative effects, relevant to both climate modelling and satellite aerosol retrieval schemes.~~ was found.

1 Introduction

Vegetation fires are estimated to emit $\sim 2.0\,\mathrm{Pg}$ of carbon per year into the atmosphere (van der Werf et al., 2010) influencing air quality, weather and climate (Bevan et al., 2009; Langmann et al., 2009). Particulate matter emissions adversely affect human health and mortality rates (Johnston et al., 2012), and have a substantial but very uncertain effect on the Earth's radiative budget (Bond et al., 2013). Climate model intercomparison (Myhre et al., 2013) indicates little agreement in simulated magnitude and sign of direct radiative forcing attributed to biomass burning emissions. Smoke plumes are difficult to observe, characterise and represent in climate models because they contain chemically and microphysically complex particles that evolve during their atmospheric lifetimes. Improving the characterisation of emission factors, aerosol optical properties, mixing effects and microphysical processes in modelling schemes were identified as key directions narrowing the uncertainties and reducing model and observational biases (Koch et al., 2009; Bauer et al., 2010; Bond et al., 2013).

Characterisation of smoke aerosols is a fundamentally difficult problem because of the dynamic nature of combustion particle formation and evolution. Particulate emission from open biomass burning consists ~~primarily of scattering organic matterand soot – agglomerates of carbon nanospheres (?) which are strong absorbers and have a significant warming effect (Bond et al., 2013) . Soot structures~~ of organic matter, soot carbon and trace inorganic elements. Different chemical species are not discretely separate, but are ~~embedded in, or have organic coatings~~ internally mixed (Reid et al., 2005a). Smoke particle size distributions are typically bimodal with the bulk of mass concentrated in the fine mode (aerosols 0.1 to 1 µm in diameter). ~~The~~ Particle production rates, the proportions of different chemical species, particle size distributions, mixing state and hence optical properties of the particles vary greatly depending on the fuel type and moisture, ~~combustion phase~~a

ratio of flaming versus smoldering combustion, fire intensity and meteorological conditions at the time of emission (Reid et al., 2005a, b; Janhäll et al., 2010, and references therein).

Smoke emitted from different vegetation types indicate distinctiveness in particle size distributions and optical properties. Emissions from Northern temperate and boreal forest fires are generally less absorbing and tend to have larger particles than Amazonian or African forest burn plumes (Hobbs et al., 1996; Dubovik et al., 2002; Eck et al., 2003) , due to differences in fuels, combustion phase and fire intensity (Reid et al., 2005a) . Flaming fires are generally thought to emit larger and more absorbing particles (Hobbs et al., 1996; Reid and Hobbs, 1998; Janhäll et al., 2010) . However, there is evidence that strongly smouldering combustion of peat fuels in particular can generate even larger particles (Nakajima et al., 1999; Eck et al., 2003) .

The complexity of the parametrisation increases with ageing processes. Emitted smoke particles are lofted to altitudes ranging from hundreds to thousands of meters, frequently above the planetary boundary layer (Kahn et al., 2008; Martin et al., 2010), and in some cases to the lower stratosphere (Damoah et al., 2006). During the lifetime of several days to weeks plumes can be transported on regional (Colarco et al., 2004) or intercontinental (Damoah et al., 2004; Dirksen et al., 2009) scales. Ageing aerosols undergo chemical and physical transformations. Measured increases in particle size have been attributed to coagulation (Reid et al., 1998; Fiebig et al., 2003; Colarco et al., 2004) , condensation (Cocker et al., 2001; Carrico et al., 2005) and coating (Dahlkötter et al., 2013) . The exact effects of these processes on the radiance field are not established and only recently has ageing been introduced into modelling schemes (Bauer et al., 2010) . An increasing number of studies indicate the impact of aerosol mixing state and morphology (Bauer et al., 2010; Shiraiwa et al., 2010; Cappa et al., 2012) on optical properties stressing the need to parameterise these processes in models. A

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 µm 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) . I addition, water uptake by particles can be a significant, in particular at higher relative humidity and for particles with higher amounts of soluble material. Inferred hygroscopic growth factors suggest an increase in particle size by up to ~15% (Magi and Hobbs, 2003; Rissler et al., 2006) . The above particle growth processes enhance smoke scattering efficiency. A number of studies report increases in a ratio of scattering extinction to total extinction, single scattering albedo (SSA(λ)) ~~, has been reported to increase~~ in ageing plumes ~~(Reid and Hobbs, 1998; O'Neill et al., 2002; Abel et al., 2003; Eck et al., 2009) indicating greater enhancement in scattering efficiency compared to absorption. In addition, laboratory based experiments (Shiraiwa et al., 2010) and modelling studies (Jacobson, 2001; Bauer et al., 2010) suggest a significant increase in absorption in ageing smoke due to the lensing effect, where by the thickening coating of scattering matter increases particle optical cross-section and redirects more radiation towards the absorbing core. Ambient aerosol measurements (Cappa et al., 2012) , however, indicate only modest absorption enhancement~~(O'Neill et al., 2002; Abel et al., 2003; Eck et al., 2009) .

Northern temperate and boreal forest fires generally are large and intense, with significant contribution of smoldering combustion. Plumes tend to be less absorbing with larger particles than African savanna or Amazonian forest emissions, and exhibit large variability e in retrieved properties (Dubovik et al., 2002; Eck et al., 2003) . Inferred large particle sizes could be attributed to significant smoldering phase burning, inefficient

combustion in very intense flaming fires and accelerated coagulation rates in highly concentrated plumes (Hobbs et al., 1996; Reid et al., 2005a) . Even larger particles, with R_{fv} of ~0.25 µm, have been observed in aged plumes originating from strongly smoldering combustion of peat fuels (Nakajima et al., 1999; Eck et al., 2003, 2009) .

Observation and retrieval of smoke ~~optical parameters~~ particle properties by remote sensing is challenging because of particularly high spatial and temporal variability in plume occurrence and evolution. Satellite remote sensing provides global and continuous measurements of Aerosol Optical Thickness (AOT) with improving accuracy and between-sensor agreement (Kinne et al., 2003; Kokhanovsky et al., 2010; de Leeuw et al., 2013; Holzer-Popp et al., 2013). However, while methods based on UV absorption, or multi-angle retrieval offer potential to resolve further aerosol properties, (Torres et al., 2007; Kahn et al., 2009; Dubovik et al., 2011), the presence of variable background reflectance over land surfaces makes routine operational retrieval of aerosol absorption problematic, especially at low optical thickness. Accordingly, many retrieval schemes make use of a priori knowledge of properties of characteristic aerosol types within retrievals.

Ground based Aerosol Robotic Network (AERONET) sun and sky photometers (Holben et al., 1998) located at locations worldwide, provide more robust aerosol measurements. The observations are less affected by the surface reflectance component and are not limited to one or a few view angles. Retrieved aerosol optical and physical properties are fundamental in determining dominant aerosol types for various regions (Dubovik et al., 2002; Lee et al., 2010; Giles et al., 2012) and have become a benchmark for validating satellite observations (Kokhanovsky et al., 2010; Holzer-Popp et al., 2013). AERONET aerosol characterisations have been derived by taking average values from a set of stations assumed to be representative to a certain region or aerosol type. This approach approximates complexities and variability in aerosol properties, but it does not exploit some of the information contained in individual AERONET observations. For example a record from stations located at or near boreal forest is likely to include observations of smoke of various age and origin. Several studies used atmospheric transport modelling and satellite data to determine the source and age for a set of AERONET smoke observations, focusing on individual burning

events (O'Neill et al., 2002; Eck et al., 2009; Dahlkötter et al., 2013). Sayer et al. (2014) subdivided sets of AERONET stations representing biomass burning regions into near source and distant ones to explore the ageing effects on optical and microphysical properties. These studies stress the need for smoke plume source and age resolved analysis methods establishing particle properties for different emission sources and long term ageing effects.

This study addresses this research gap by improving the characterisation and ageing effects of smoke plumes typically attributed to Northern temperate and boreal forests. A new method is presented here allowing the estimation of age and source for AERONET aerosol observations. The method is applied with a focus on two aims. The first is to determine smoke particle microphysical and optical properties for emissions from different vegetation types. Explicit source attribution offers additional information content compared to region-based approaches (Dubovik et al., 2002; Giles et al., 2012; Sayer et al., 2014) and could partly address the large variability in aerosol properties characteristic to the boreal forests. The second aim is to explore changes in particle properties occurring in plumes over several days of ageing, complementing existing studies ~~(Reid et al., 1998; Capes et al., 2008; Eck et al., 2009)~~ (Radke et al., 1995; Reid et al., 1998; Cap
independent estimates based on an alternative method and larger sample.

2 Data and methods

The source and age estimation for AERONET smoke observations was achieved by using an air parcel trajectory model and satellite active fire and AOT observations. For all of the selected AERONET smoke observations described in Sects. 2.1 and 2.1.1, a set of air parcel back trajectories ending at a range of altitudes was generated using the HYSPLIT model as specified in Sect. 2.3. Coinciding satellite active fire (Sect. 2.2) and AOT observations (Sect. 2.4) along the trajectories were used as inputs into the decision tree outlined in Sect. 2.5 and Fig. 4, estimating source and age for the AERONET observations.

2.1 AERONET data

The analysis is based on AERONET Level 2.0 version 2 inversion data products. AERONET CIMEL radiometers are calibrated and continuously monitored, and data products are cloud screened and undergo robust standardised processing (Holben et al., 2006) which enables quantitative comparative analysis. Direct solar extinction measurements provide columnar AOT at several wavelengths ranging from 340 to 1640 nm. Combined direct sun and diffuse sky radiance measurements at four wavelengths (440, 676, 879 and 1020 nm) are best-fitted with radiative transfer model effectively retrieving columnar aerosol size distribution, spectral complex refractive index, SSA(λ), ~~Angstrom~~ Ångström Exponent (AE), Absorption ~~Angstrom~~ Ångström Exponent (AAE), phase function and precipitable water content (Dubovik and King, 2000; Dubovik et al., 2006). Retrieved aerosol optical properties and size distributions are in agreement with independent in-situ aerosol measurements (Haywood et al., 2003; Johnson et al., 2009) and have well defined uncertainties (Dubovik et al., 2000; Holben et al., 2006). The accuracy ~~is improved during higher~~ of optical properties retrieval is increased during moderate and high aerosol loading conditions~~and therefore AERONET level~~ . Level 2 ~~inversion products contain only retrievals~~ inversions contain SSA retrievals only at AOT(440) levels of 0.4 or higher. AERONET uncertainty for individual SSA retrievals in these cases is approximately 0.03 (Dubovik et al., 2000) . Particle fine mode volume median radius has an uncertainty of 0.01 µm, the spread of the fine mode particle distribution – 0.06. Estimated uncertainty in asymmetry parameter ranges from 0.015 at AOT(440) to 0.04 at AOT(1020) (Dubovik et al., 2000; Sayer et al., 2014) . Size distribution parameters and optical properties discussed throughout this study are consistent with the definitions and units described in Holben et al. (2006).

2.1.1 Data selection

This study uses data from AERONET stations positioned within or in proximity to the Northern boreal and temperate forests. All available level 2 data with AOT(440) level ≥ 0.4 from the stations located north of 45° latitude in North America and Asia (Fig. 1) and collected at

any time from the year 2002 through 2013 were selected. European observations were excluded because of higher background AOT levels and more likely mixing of smoke plumes with urban and industrial aerosol. Temporal extent of the study was constrained by the start date of MODIS data availability from both platforms Terra and Aqua in early 2002. AOT record from the selected AERONET stations indicate generally low aerosol background levels with sharp spikes in aerosol loading occurring during the burning season lasting from late spring to early autumn. The pattern suggests that the majority of level 2 retrievals ~~(~~at AOT(440) ~~> 0.4~~levels of ≥ 0.4) is a record of biomass burning plumes. Notably, this study is not fully inclusive or exclusive to the Northern forest emissions both in terms of vegetation type or geographic extent. Plumes transported to the selected AERONET locations from areas extending beyond the region of interest and attributed to a range of land cover types have been included in the analysis.

Severe burning seasons in 2004 and 2005 in Alaska caused very high AOT(λ) values $\sim$ (2–5) recorded at Bonanza Creek AERONET station. The very large (0.2–0.25) ~~fine mode volume median radius (~~R_{fv} ~~)~~ values retrieved during these events were attributed to peat fuel combustion (Eck et al., 2009). The method used in this study could not establish ageing properties for these plumes because of large number of active fires and persistently elevated AOT levels in the region. As a result Bonanza Creek observations for August in years 2004 and 2005 have been excluded from the analysis.

2.2 Active fire data

As a proxy for fire activity during the period analysed, the MODIS fire location dataset MCD14ML produced by the University of Maryland and provided by NASA Fire Information for Resource Management System was used. The dataset contains active fire detections from Terra and Aqua platforms with information of the hot spot location, brightness temperature at MODIS bands 21 and 31, fire radiative power and detection confidence (Giglio et al., 2003).

2.2.1 Fire data processing and selection

Fire inventories compiled for Alaska (Stocks et al., 2002) and Canada (Kasischke et al., 2002) indicate that very large fires are not numerous, but account for the majority of the total area burned. Stocks et al. (2002) found that fires larger than 100 km^2 represented more than 80 % of total area burned. Following this only large wildfire events likely to be strong emission sources were considered in the analysis. To identify such fires MODIS individual hot spots with 80 % or more detection confidence were agglomerated into fire events using the Density Based Spatial Clustering (DBSCAN) algorithm (Ester et al., 1996). DBSCAN clustering was performed merging individual fire detections into a single fire object if at least two were found closer together than 10 km in space and 24 h in time. The objects were iteratively formed by adding any fire points found within the search radius from all of the fire detections belonging to the cluster (Fig. 2). Identified fire events were considered as large and selected for the analysis if the event duration was more than 48 h, and the spatial bounding box including all points belonging to the event was larger than 100 km^2.

2.2.2 Fire and emission source land cover type

The emission source land cover type for each of the fire events was determined using MODIS MCD12C1 annual land cover type data products which employ 17 different land cover classes defined by the International Geosphere Biosphere Programme (Loveland and Belward, 1997). Initially, the land cover type was identified for each of the active fire pixels within a fire event from a grid value given in the MCD12C1 product from a corresponding year. The land cover value occurring most often (mode) was used as a land cover type identifier for the fire event. This was done for all of the years except 2013 for which MCD12C1 is unavailable and the 2012 product was used instead.

2.3 Back trajectories

To link AERONET observations with source regions and to identify the likely smoke transport pathways, air mass trajectories were computed with HYSPLIT (Hybrid Single-Particle

Lagrangian Integrated) model (Draxler and Rolph, 2003). The HYSPLIT model was run using Global Data Assimilation System (GDAS) meteorological archive data for the available 2005–2013 period and NCEP/NCAR reanalysis data for 2002–2004. For each of the studied AERONET elevated AOT observations (AOT at 440 nm above 0.4) seven day back trajectories with one hour temporal step were generated starting at 16 elevations ranging from 500 to 12 000 m: at 500 m intervals below 4000 and at 1000 m intervals above 4000 m. The uncertainty in the individual trajectories was assessed, estimating HYSPLIT Model numerical integration and meteorological data resolution errors. The first was estimated computing back trajectory and then forward trajectory from the back trajectory's end point. The error was assumed to be half of the horizontal and vertical distance between the initial start and the final end points. The resolution error and resultant divergence in flow field was determined generating a grid of 27 ($3 \times 3 \times 3$) back trajectories beginning around the initial start point, with horizontal and vertical offsets given by the estimated numerical error.

2.4 Satellite AOT

The two independent satellite AOT data products used are based on observations from (1) The Moderate Resolution Imaging Spectrometer (MODIS) sensors on-board Terra and Aqua platforms and (2) Along Track Scanning Radiometer (AATSR) sensors flown in succession on ERS-2 and ENVISAT spacecrafts. While the method can readily be extended to include further satellite data, the role here is to confirm model tracking of plume transport from source to AERONET, rather than to add additional information on aerosol properties. The MODIS colection 5.1 dataset M*D04_L2 is based on the dark target retrieval scheme (Kaufman and Tanre, 1998b; Levy et al., 2009) and AATSR_SU on the algorithm developed at Swansea University (North, 2002; Bevan et al., 2012), modified under the ESA Aerosol Climate Change Initiative (CCI) (Holzer-Popp et al., 2013; de Leeuw et al., 2013). Both data products provide interpolated AOT at 550 nm ~~and have~~ with 10 km $\times$ 10 km ~~spatial resolution. The MODIS dataset has greater spatial coverage and temporal resolution because of wider swath~~ pixel size at nadir, which is doubled at the edge of swath for MODIS and increase only negligibly for AATSR. Because of the wider view angle and two sensors operational

at the same time~~. Importantly, both~~ , The MODIS dataset has greater spatial coverage and temporal resolution. The algorithms perform well compared to AERONET AOT observations. Validation studies suggest RMSE from 0.1 to 0.2, and little bias between AERONET and both satellite AOT retrieval schemes (Levy et al., 2010; Holzer-Popp et al., 2013). ~~It must be noted that the assessments~~ The product validations and RMSE as a ~~measures themselves~~ measure are biased towards low AOT conditions, which constitute the vast majority of the observations. Discrepancies ~~can be expected to be~~ are larger for the aerosol loadings (AOT(440) ~~> 0.4~~ ≥ 0.4) analysed herein. ~~However, the accuracy of satellite AOT observations was not critical for the purpose of this study~~Estimated MODIS AOT one sigma expected error bounds increase linearly with AOT $\pm(0.05 + 0.15\%AOT)$. AATSR SUBSCRIPTNBSU algorithm retrieves AOT only up to 2.0, meanwhile MODIS M*D04 SUBSCRIPTNBL2 product contains AOT values up to 5.0. Notably, 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) .

2.5 Age and source estimation

Smoke source attribution for AERONET observations was performed by finding coinciding satellite AOT and fire event observations along the generated trajectories and identifying candidate plume pathways. Starting at an AERONET station and an observation time, for each trajectory level spatiotemporal queries were performed, finding any satellite AOT and fire event observations falling within the trajectory domains at hourly steps (Fig. 3). The location and size of spatial search windows at each time step was given by the trajectory uncertainty analysis. Identified AOT observations and proximity to large fire events served as inputs into the decision tree. A trajectory was selected as a candidate if the conditions showed in Fig. 4 were satisfied. When several candidate trajectories were selected, the trajectories were ranked according to the potential fire source size and satellite AOT values observed after the trajectory has passed the source. Finally, the highest ranked candidate was identified as the source and age estimate.

2.6 Statistical methods

~~A linear regression was used to examine ageing effects on smoke particle microphysical and optical properties. Both positive or negative correlation between the age estimates and a particular property was considered significant if two-sided p value was ≤ 0.01. Error estimates and uncertainty intervals~~ Error bars and uncertainties on the quantities stated represent 95 % confidence intervals ~~in regression coefficients.~~

~~The differences in properties of plumes attributed to different land cover types were assessed by the Mann-Whitney U test. Two samples~~ derived using the bias-corrected non-parametric bootstrap methods (Efron, 1993). During the resampling error was modelled as normally distributed AERONET uncertainty given as 1 standard deviation. The medians of two populations are identified as significantly different if the ~~two-sided p value was ≤ 0.01~~bootstrapped 95 % confidence interval on difference in medians agree in sign.

3 Results and discussion

From a total of 1337 AERONET observations processed, age and source were determined for 629. The majority of the identifications are for smoke of up to 1–2 days of age, and ~~the counts are increasingly smaller for older estimates~~ only 6% are for plumes older than 4 days (Fig. 1c). ~~Due to a small number of identified observations of plumes aged more than 4~~The method employed was limited to tracking of highly concentrated free tropospheric plumes emitted from fires larger than 100 ~~,~~ km^2 and the results are ~~more robust when considering the first 4of ageing. This is an expected outcome because it is progressively less likely that the plume evolution is tracked successfully, satisfying all of the decision tree conditions, as smoke ages. Inclusion of European AERONET observations could potentially increase the numbers for old smoke as many aged North American and, to a lesser extent, Asian boreal forest plumes are observed in Europe (Damoah et al., 2004; Dahlkötter et al., 2013)~~ 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. However, ~~the method currently does not perform well in the cases of extremely long transport and high background AOTlevels, and thus the number of additional estimates would be small~~fire inventories (Stocks et al., 2002; Kasischke et al., 2002) indicate that large burning events is the dominant mode of burning in boreal and temporal forests. The results of this study suggest that the large plumes are accountable for at least 45% of AERONET AOT(440) $\geq$ 0.4) observations in the region.

3.1 Emission source

The most frequent emission source land cover type determined was North American evergreen needleleaf forest (ENF) constituting more than half of all estimates (Fig. 1b). The dominance could indicate ~~exceptional fire activity or emission characteristics resulting in significantly more plumes originating in ENF compared to other land cover types. However, we suggest that it is likely to be merely a consequence of ENF spatial distribution and location~~ larger and more intense fires characteristic to North American forests compared of Eurasian boreal regions (de Groot et al., 2013) , but this may be caused by uneven spatial distribution of the AERONET stations~~used in the analysis. ENF occupies large areas stretching from North to South in Western Canada and USA with many AERONET stations located to the East. Smoke dispersion towards the stations is accommodated by prevailing East–West transport at these latitudes. It is important to consider sample size inconsistencies when evaluating the variability in particle properties between plumes attributed to different land cover types~~. Emissions attributed to woody savannas and open shrubland were split between the continents. All mixed forest plumes represent Eurasian fires. Sample size inconsistencies and uncertainty in source attribution should be considered evaluating the results. Distributions for classes with a small number of estimates exhibit multimodality (Fig. 5) and thus their summary metrics should be treated with caution. 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 $\sim$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. Despite these limitations the emerging patterns suggest meaningful differences between the plumes attributed to the land cover types discussed.

Plumes show variability in particle concentrations, as indicated by the maximum satellite AOT value along the determined 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. If this interesting feature is not introduced by sampling biasses or satellite AOT retrieval artefacts, it may indicate two distinct emission modes.

Grassland and cropland emissions are distinctively less optically thick, in agreement with observations that flaming grass and shrub fires are less intense and produce less particles than forest fires (Reid et al., 2005a) . Notably, Eurasian mixed forest plumes are predominantly in the extremely concentrated group, open shrubland and wooded savannas emissions shared between the continents show clear bimodality, meanwhile North American ENF plumes are primarily less optically thick with smaller proportion of very dense plumes. Satellite observations and fire inventories suggest that North American fires tend to be high intensity crown burns consuming more fuel per area burned than predominantly surface fires in Eurasian boreal forests. (Wooster and Zhang, 2004; de Groot et al., 2013) . The apparent higher proportion of very dense plumes in Eurasia in this study might be caused by the small number of AERONET stations in the continent and thus greater sampling bias. However, our data also indicates that the extreme events tend to occur north of 50° latitude, and in biomes with lower tree cover density. Thus it seems unlikely

that such events can be explained solely by variability in intensity of crown and surface fuel combustion. The highest proportion of total fuel in boreal regions is contained below ground as organic soils, peat and root material (van Leeuwen et al., 2014) . Notably, the highly dense plumes attributed to mixed forest fires include well documented (Chubarova et al., 2012) forest and peatbog fires in Russia in year 2010. During the events AERONET AOT(500) values as high as 6.4 were recorded in Moscow. This suggests that ground fuel combustion triggered at least some of the 165 extremely concentrated plumes discussed. Such events are probably overrepresented in this study, in particular in Eurasian continent. Nonetheless they comprise at least 12 to 25% of all AERONET(440)$\geq$ 0.4 observations in the region and period studied and require further investigation.

Particle ~~size distributions derived for the source~~ distributions of smoke originating from the land cover type ~~classes~~ fires discussed indicate distinctiveness in fine mode volume median radius ~~(~~and spread of R_{fv}~~)~~. Cropland/natural vegetation mosaic and grassland emissions tend to show the smallest particles, with median R_{fv} values 0.143 ~~and 0.147~~(0.135–0.145) and 0.157 (0.148–0.168) µm. Plumes from mixed forests generally contain the largest particles followed by ~~wooded savannas~~ open shrubland emissions having median R_{fv} values ~~0.193 and 0.186~~0.194 (0.184–0.2) and 0.185 (0.176–0.194) µm respectively. The most numerous ENF smoke observations typically have particles smaller than the emissions from mixed forests and woody savannas, with determined median R_{fv} 0.164 (0.16–0.167) µm. Significant difference in R_{fv} exists comparing less dense and very dense plumes, estimated median R_{fv} are 0.163 (0.16–0.166) and 0.191 (0.184–0.195) µm for the two initial concentration categories. The width of the fine mode particle distribution is positively related to R_{fv}, and classes with larger particles have wider distributions.

These estimates generally agree with published size distributions for various fuel types. Grass fires are reported to emit smaller particles (Reid and Hobbs, 1998) than forest fires (Dubovik et al., 2002). ~~Plumes with the smallest median R_{fv} in this study are attributed to grass, cropland/natural vegetation mosaic and ENF fires. This could be because of smaller fire intensities in these cover types compared to fires in mixed forest and wooded savannas (Reid et al., 2005a) . As well as differences in burning intensity, large particles in the later~~

classes could be influenced by the presence of significant amounts of peat in these biomes, the combustion of which is known to generate very large particles (Eck et al., 2009) .

Boreal smoke particles are known to be generally larger R_{fv} compared to African or Amazon forest emissions (Reid et al., 2005a) , but also exhibit high variability in retrieved properties (Eck et al., 2003, 2009) . The distinctiveness in particle size distributions attributed to the three forest cover types – ENF, mixed forests and wooded savannas – is particularly interesting. It indicates significant differences in fuels consumed, combustion conditions and fire intensities between these vegetation types which are typically agglomerated under broad boreal forest definition. Boreal smoke size distributions are known to have generally larger extra-tropical or boreal forest definitions. Differences in median R_{fv} compared to African or Amazon forest emissions (Reid et al., 2005a) , but are also reported to exhibit high variability in retrieved properties (Eck et al., 2003, 2009) . The results obtained in this study indicate that could be partly explained by varying proportions of very dense plumes attributed to these cover types. Wooded savannas, open shrubland and, in particular, mixed forest emissions are dominated by such events. Inferred particle sizes in optically thick plumes are close some of the variability could be addressed by the source attributionlargest documented values (Eck et al., 2009) , suggesting that they might be produced by smoldering combustion of peat fuels. Accelerated coagulation rates in highly concentrated plumes and higher particle hygroscopisity may increase R_{fv} even further. Smaller particles in ENF could be due to predominantly flaming combustion of crown fuels, inferred lower initial concentrations and lower coagulation rates just after emission.

Notable differences exist in fine mode particle distributions comparing daytime and night-time emissions. Fire radiative power observations indicate a strong diurnal cycle in burning intensity with a peak at around 14 – 16pm local time (Ellicott et al., 2009) . Results obtained by this study imply that plumes emitted between 12pm and 22pm local time tend to have larger particle fR_{fv} than the ones emitted between 24pm and 8am (Fig. 5e). Plumes aged between 12 and 48 hours were compared, expecting higher accuracy in age estimates for such observations, and minimizing very young smoke bias and long term ageing effects. Although the spread is large and overlaps are substantial, the day and night differences

in R_{fv} are significant for the less concentrated plumes. Daytime and plumes generally have larger particles, with a median R_{fv} value of 0.17 (0.167–0.18) µm, compared to 0.155 (0.148–0.161) µm value for night emissions. A very small difference exists in SSA(440) values. Day plumes are only insignificantly more absorbing, estimated median SSA(440) values are 0.948 (0.943–0.956) and 0.956 (0.952–0.962) for the day and night emissions respectively. Reid et al. (1999) report larger and less absorbing particles produced at night in Brazil attributed to smoldering combustion. Different diurnal particle size pattern suggests that larger and more intense flaming daytime fires in boreal regions generate larger particles then more smoldering night–time burning.

The differences in optical properties between plumes attributed to the land cover classes considered are ~~more~~ subtle. The variability in single scattering albedo and its spectral dependence is small. Inferred median SSA(440) is ~~within the AERONET uncertainty interval of 0.02 from an~~ close to often reported values of 0.95 ~~value~~ for boreal regions (Dubovik et al., 2002; Reid et al., 2005b) for all land ~~covers~~ cover types except for the plumes from cropland/natural vegetation mosaic fires, which are more absorbing and have a median SSA(440) value of 0.9 (0.886–0.916). Notably, in this land cover class the lowest SSA(440) corresponds ~~both~~ to the smallest median R_{fv} ~~and to the highest median imaginary part of the refractive index~~ (Fig. ~~7a, d and b~~5). Smoke from mixed forests, with a median SSA(440) value of 0.963 (0.956–0.977), is slightly less absorbing than ENF~~and wooded savannas emissions which both~~ , wooded savannas and open shrubland emissions which all have median SSA(440) values ~~of~~ not significantly different from 0.95. These differences in absorption magnitude suggest some variability in flaming vs. smouldering combustion ratios and is consistent with observed differences in size distributions. However, there is little to differentiate between the different forest types considered here based solely on optical properties. ~~The often reported SSA(λ) values close to 0.95 for boreal regions (Dubovik et al., 2002; Reid et al., 2005b) seems to be applicable for emissions from all forest types discussed here.~~

3.2 ~~Particle growth rates~~ Ageing effects on particle distributions

~~There is a significant relationship between the age estimates and AERONET retrieved smoke~~ Significant differences exist comparing AERONET retrieved particle size distributions of young and well aged plumes. Figure 6a shows ~~AERONET retrieved~~ fine mode volume median radius (R_{fv}) plotted against the age estimates. ~~A linear fit suggests a R_{fv} growth rate of 0.0084~~Notably, less dense plumes dominate young estimates, while optically thick large plumes are more frequently detected after several days of ageing. Because of the sampling bias and inherent large differences in size distributions between less dense and very concentrated plumes, the ageing effects for the two smoke categories are compared separately. Low number of very old estimates only allow meaningful comparison of plumes of up to 4 ~~(0.0072~~days old, and estimates older than 72 hours were pooled together. Distributions of plumes binned into groups per day of ageing indicate a steady increase in median R_{fv} for the first 3 days, and only slightly larger particles for plumes aged for more than 3 days. A similar pattern is observed for both plume density categories. Plumes in the oldest age category have median R_{fv} values larger by 0.02 (0.009–0~~.0098~~.028) and 0.022 (0.012–0.036) µm compared to young smoke of up to a day old for the less dense and very dense plumes respectively. This difference would equate to a ~0.007 (0.003–0.012) µm flat growth rate in median R_{fv} per day.

The trajectory analysis indicates that in several cases the same plume was transported over more than one AERONET station allowing to infer changes in plume properties between the two observations. Unfortunately, only 13 of such events were identified preventing a more robust analysis (Fig. 6a). In 10 out of the 13 cases older particles are larger, while three of the pairs suggest a decrease in R_{fv} between the observations. The median R_{fv} change rate is 0.0075 (-0.001–0.03) µm ~~CI)~~ per day. The estimate agrees well with the growth rate suggested by differences in particle distributions between young and aged plumes. However, it has large confidence intervals due to low number of paired observations and uncertainty in individual AERONET retrievals.

Published smoke particle growth ~~rates~~ magnitude and time scales vary and are difficult to compare due to differences in measuring time interval, techniques, sampled fuels, fire size, intensity, combustion phase and smoke age. Rapid increases by as

much as 0.08 µm have been reported (Hobbs et al., 1996; Abel et al., 2003) , suggesting instantaneous R_{fv} growth rates of ~0.04 µm per hour. Reid et al. (2005a) concluded in their review that ~~, on average,~~ aged smoke particle distributions typically have R_{fv} larger by 0.025 µm. ~~In agreement with this, the growth rate derived~~ Importantly, most of the particle growth is attributable to condensation and coagulation happens during the first few hours. 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. The changes in particle size distributions inferred in this study ~~indicates an increase in R_{fv} of 0.025in approximately 3~~happen over the course of days, suggesting that in thick continental plumes particles continue to grow. The principle mechanism driving size increase on such time scales is thought to be coagulation (Radke et al., 1995; Reid et al., 1998; Capes et al., 2008) .

The ~~SD from~~ spread of R_{fv} does not exhibit significant dependence on estimated age. Generally, distributions with smaller R_{fv} tend to have a ~~smaller spread~~ narrower width (Fig. 6~~d~~b). The relationship between R_{fv} and ~~R_{fv} spread~~fine mode spread, however, is stronger for young smoke and is not evident for the aged smoke observations. This indicates that while particle sizes increase in ageing plumes, the spread of the fine mode volume radius does not change systematically.

~~The age and R_{fv} relationships seen in Fig. 6a exhibit a degree of non-linearity. Quadratic fit indicates initially faster and gradually decreasing growth rates. Due to the small number of very old smoke observations it is not possible to determine whether the non-linearity is significant. However, the data available suggests that there is a change in the R_{fv} increase trend after 3–4of ageing. Notably, a linear fit on the selected data subset of observations up to 4old indicates a higher aerosol growth rate of 0.0095(0.0069–0.012CI) per day. In that case 0.025in R_{fv} is gained in between 2 and 3. The inferred gradual slow down in growth integrates well with the reported stabilisation in size distributions in Brazilian smoke after 3of ageing (Kaufman et al., 1998a; Reid et al., 1998) . These studies suggest the increase in size is~~ Turco and Yu (1999) suggested that condensation narrows the particle distribution,

while growth due to ~~coupled coagulation and condensation processes during the first 1–4followed by predominantly coagulationgrowth afterwards~~coagulation does not alter the width significantly. The lack of significant changes in fine mode spread suggests that the inferred particle growth is attributable to coagulation.

~~The derived linear model coefficients are significant but the spread of the values is large. The high variability can be partly explained by significant differences in size distributions between plumes attributed to different land cover sources. The growth rates derived for smoke of different origin seen in~~

3.3 Changes in optical properties

The Ångström exponent (AE) is tends to be lower for older plumes (Fig. 7~~a are notably similar despite varying sample sizes. All estimates fall within the confidence interval of the regression slope for all of the data with the exception of ENF plumes for which the derived growth rate is slightly lower.~~

~~Variances in particle R_{fv} within the fire source classes are nonetheless large. This could be due to any of the following factors: (1)erroneous age and source attribution in the case of mixture of fresh smoke and aged regional haze or mixture with different aerosol species; (2) inevitably large intra-class variability in a range of factors determining particle sizes during the combustion; (3) fast processes beyond the temporal resolution of the proposed method and data. A number of studies have measured very rapid dynamics including significant particle growth during the first few hours of ageing (Hobbs et al., 1996; Abel et al., 2003; Calvo et al., 2010) . Growth rates can be particularly high in concentrated plumeswith high aerosol loading due to more frequent collisions between particles, fast condensation (water and organics) and gas to particle conversion. The AERONET records typically do not include observations of truly fresh smoke within seconds or minutes after the emission. As a result of this limitation and the uncertainty originating from the age estimation method employed in this study, the analysis has only a moderate temporal resolution and is limited to relatively long term ageing processes. Figure 6a indicates that fresh smoke observations with largest retrieved R_{fv} tend to have~~

higher AOT values and wider R_{fv} spread. Assuming that the age and source estimates are correct for these cases, the explanation may be unusual fuel and combustion characteristics or fast particle growth at high aerosol concentrations.

3.4 Changes in precipitable water content

There is a significant correlation between AERONET retrieved columnar precipitable water content and the age estimates (Fig. 7c). Generally, older plumes tend to be mixed in air masses with higher water content. The relationship is similar to R_{fv} ageing pattern, but suggests an even higher degree of non-linearity. The quadratic fit exhibits a peak in AERONET precipitable water content at around 3–4of ageing and a subsequent decrease. This pattern suggests that, when plumes are being transported further from sources, they are entrained within progressively more moist air masses. A relationship between water vapour content and AOT has been observed (Hegg et al., 1997; Smirnov et al., 2000) , indicating that it is one of the main contributors to total AOT. The results presented in this study show that plumes from mixed forest and open shrubland b). In well aged very dense plumes median AE changes by -0.15 (-0.036 to -0.26) from a 1.74 (1.64 – 1.82) value for young plumes. The difference is smaller and not significant for the less dense plumes, for which median AE changes by -0.06 (0.002 to -0.15) from median value of 1.89 (1.86 – 1.92) estimated for young plumes. Decreasing wavelength dependence of AOT corresponds to inferred particle growth and is an expected result. Changes in median AE, however, are not as consistent as differences in median R_{fv} between the plume age and density categories. This variability could be due to uncertainties in AERONET inversions, insufficient sample size for some land cover classestend to be transported within air masses with higher water vapour content. Notably, the same plumes tend to have larger particles and higher growth rates. This may suggest that water condensation plays a role in inferred particle growth during the first few days of ageing, complementing reported transition from condensation and coagulation growth to coagulation growth after 3–4of ageing (Kaufman et al., 1998a; Reid et al., 1998) , or they could reflect effects of differences in particle chemistry affecting AE besides particle size.

3.4 ~~Changes in optical properties~~

In contrast to particle ~~size distributions, the~~ particle size distributions and AE, differences in SSA(λ) ~~and age relationship is not statistically significant and falls within the uncertainty interval of 0.02 for individual AERONET~~ are only subtle. SSA(λ) ~~retrievals (Fig~~is only slightly higher in plumes aged for more than 3 days compared to young smoke (fig. 7~~e)~~ c). The variability is negligibly small between the two plume density categories. Combined median SSA(440) insignificantly increases from to a ∼0.95 value for young smoke to ∼0.96 for the oldest plumes. Mie theory predicts that particle radius growth due to coagulation should generally result in smaller absorption and higher scattering efficiency, hence an increase in SSA(λ) (Bond and Bergstrom, 2006). Such an increase in ageing smoke has been observed in Brazil (Reid et al., 1998), West Africa (Abel et al., 2003; Capes et al., 2008), Spain (Calvo et al., 2010) and North America (Eck et al., 2009). Notably, most of the aforementioned studies were analysing inherently more absorbing smoke with typically smaller particles compared to emissions from the sources explored in this study. It is possible that most of the scattering enhancement happens during the rapid evolution phase within few hours from the emission and thus can not be observed by the method employed in this study. Cropland/natural vegetation plumes are significantly more absorbing, but with only 34 estimates a meaningful analysis of ageing effects is not achievable.

~~There is a significant tendency for the absorption angstrom exponent (AAE) to decrease at a rate of −0.034 (−0.014 to 0.054 CI) per day with age, suggesting some changes in chemical composition in the ageing plumes (Fig. 7d). The significance of condensation of organics as well as water vapour in aerosol growth is well established (Reid et al., 2005b; Dahlkötter et al., 2013) . The effects of these processes on optical properties are less clear (Bond and Bergstrom, 2006; Cappa et al., 2012) . Enhancements in wavelength dependent absorption due to increasing organic carbon content should result in higher absorption angstrom exponent values. However, the results imply a decrease in AAE in ageing plumes.~~

The angstrom exponent (AE) decreases with age (Fig. 7c) at a rate of −0.05 (−0.038 to −0.063 CI) per day considering all land cover types. Decreasing wavelength dependence of AOT corresponds to inferred particle growth and is an expected result. Similarly, the asymmetry parameter tends to increase in ageing The asymmetry parameter is higher in well aged plumes (Fig. 7f) indicating increasing d) indicating higher back-scatter due to growth in larger R_{fv}. There are, however, some notable differences between the land cover types. Estimated change rates in the asymmetry parameter and AE vary by a factor of two or more. Although they largely agree, changes in AE and the asymmetry parameter do not mirror inferred particle growth rates exactly. This variability could be due to uncertainties in AERONET inversions, insufficient sample size for some land cover classes, or they could reflect effects of differences in particle chemistry affecting AE and the asymmetry parameter besides particle size.

There is a significant negative correlation between AOT(440) and estimated plume age (Fig. 7g). AOT(The change is consistent across the wavelengths and is simmilar between the smoke concentration groups. For combined plumes at 440) values decline at a rate of −0.048 (−0.024 to −0.072 CI) per day considering all observations. The AOT(λ) and estimated age relationship is not significant for longer wavelengths . This suggests that a positive correlation between particle size and AOT (Reid et al., 1998; Sayer et al., 2014) is counteracted by the reduction in particle concentrations in ageing plumes nm the median asymmetry parameter is higher by 0.038 (0.027–0.055) in well aged plumes. At longer wavelengths the asymmetry parameter is lower, but the difference in median values between the young and aged plumes is larger.

4 Conclusions

This study presents an analysis of ageing effects on smoke particle size distribution and optical properties in regions associated with large variability in aerosol characteristics. A new method was developed allowing the source and age attribution for AERONET aerosol ob-

servations, and applied to data from stations located in or near Northern temperate and boreal forests in North America and Asia.

The results show that plume properties vary with the determined source vegetation type as defined by the land cover classification scheme. Notable variability exists not only when comparing emissions from grasslands, croplands and forests, but also different forest types. Plumes from mixed forests generally are the most concentrated and contain the largest particles with a median R_{fv} 0.193 µm, followed by emissions from wooded savannas and shrubland. Smoke attributed to evergreen needleleaf forest fires has lower initial concentrations and exhibit smaller particles than other forest emissions, having median R_{fv} value 0.164 µm which is close to the R_{fv} values of 0.143 and ~~0.147~~0.157 µm observed in plumes from cropland and grass fires. These differences appear to be partly influenced by frequent occurrence of extremely concentrated plumes in some land cover types. Such events tend to have very large particles, and at least in some cases originate from peat fires.

Optical properties do not exhibit significant variability. Estimated median SSA(440) values range from 0.95 to 0.97 for all of the sources considered in this study with the notable exception of cropland and natural vegetation plumes which are substantially more absorbing with SSA(440) value of 0.9.

~~Derived particle~~ Plumes older than 3 days have higher median R_{fv} ~~growth rates are 0.0095~~values by ~0.02 µm ~~(0.0069–0.012~~compared to smoke aged for less than a day. This suggests a ~0.007 µm ~~confidence interval) per day~~ increase in R_{fv} for the first ~~4and 0.0084(0.0072–0.0098) per day for 7~~three days of ageing. ~~These independent estimates are based on a large sample, compare favourably with existing estimates and refine growth rates obtained by different methods. The particle growth rates derived for plumes from different vegetation types are remarkably similar, implying comparable processes driving the growth. Inferred slow down in growth rates at around 3–4~~Median growth rate derived from the 13 cases when the same plume was observed at two AERONET stations is remarkably similar (0.0075 ~~of ageing has been observed before (Kaufman et al., 1998a; Reid et al., 1998) and suggests a shift in particle growth processes.~~

~~A significant relationship is observedbetween AERONET columnar precipitable water content and estimated age. It suggests that ageing plumes are entrained in air masses with higher water content. Particle growth rates are higher for smoke which is transported within air masses containing more precipitable water, implying higher contribution of hygroscopic growth~~µm). However it is not significant and highly sensitive to uncertainties in AERONET R_{fv} retrievals. No significant shift in fine mode spread in well aged plumes is observed, suggesting that the growth is driven by coagulation.

In contrast to size distributions, smoke SSA(λ) do not ~~exhibit strong dependence on age. The inferred changes in SSA(λ) are not significant and do not exceed AERONET uncertainty bounds. The angstrom exponent is decreasing~~ differ significantly with plume age. Well aged plumes are only slightly less absorbing. The Ångström exponent is lower while the asymmetry parameter is ~~increasing with age~~higher in older plumes, reflecting the increase in particle R_{fv}. ~~The absorption angstrom exponent tends to decrease with age.~~

The method and results presented here allow the microphysical and optical characterisation of particulate emissions from different vegetation types to be improved. Estimated ageing effects provide information on long term particle evolution and active processes ~~that can be used in parameterising and validating modelling schemes and improving aerosol models used by satellite retrieval algorithms~~in large and dense plumes. These independent estimates are based on a large sample, compare favourably with existing estimates and refine growth rates obtained by different methods.

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site. AATSR_SU version 4.2 datasets were made available under the ESA Aerosol CCI project http://www.esa-aerosol-cci.org.

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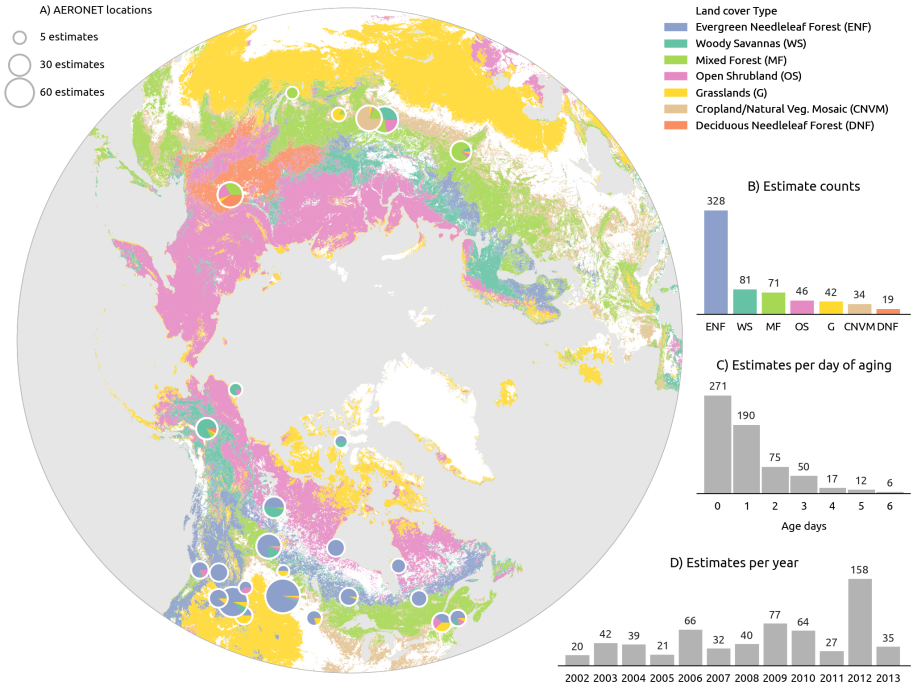

Figure 1. **(a)** Study area, land cover types examined and locations of AERONET stations used. MODIS MCD12C1 land cover data products were employed with classes defined by the International Geosphere Biosphere Programme classification system (Loveland and Belward, 1997). Pie charts inside the circles indicate the origin land cover type of plumes observed at the AERONET stations as estimated in this study. **(b–d)** show total estimate counts per source land cover type, smoke age per day of ageing and estimate number for each year.

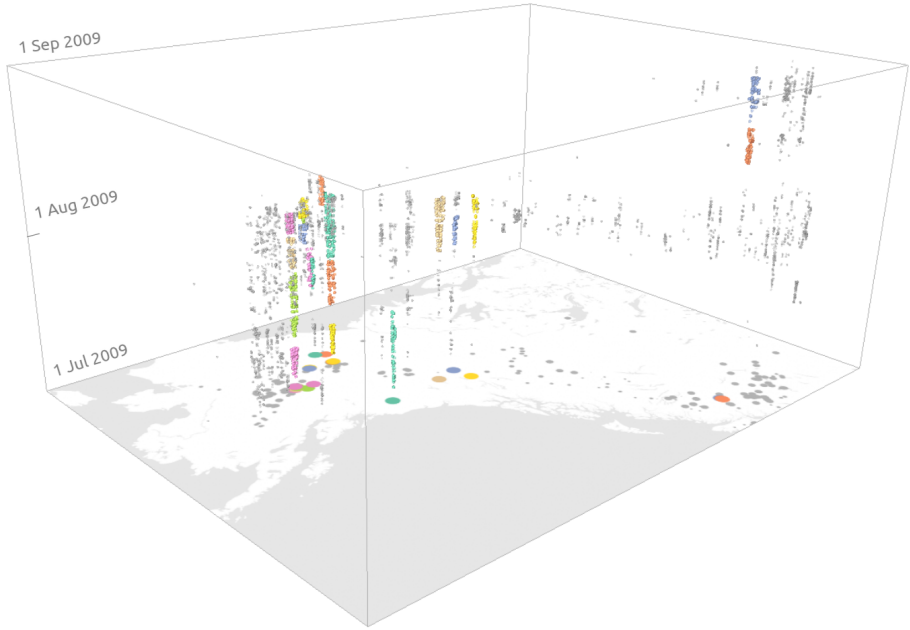

Figure 2. An illustration of the DBSCAN fire pixel segmentation method. Individual MODIS active fires in north-west Canada, Alaska and the north-western USA observed during July–August (vertical axis) in 2009. Colours show separate, large fire objects used in the analysis. Grey fire pixels were removed as described in Sect. 2.2.1.

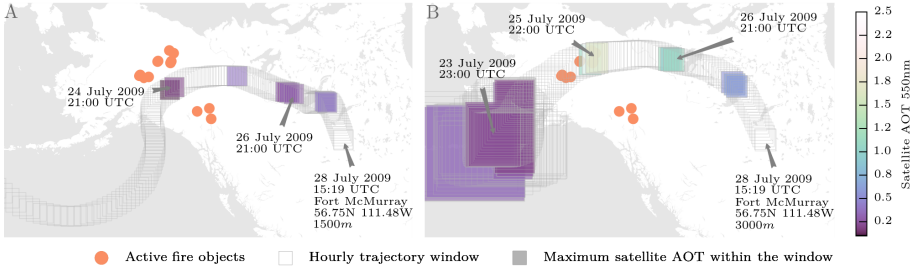

Figure 3. Two 7 day back trajectories ending at Fort McMurray AERONET station and coinciding fire and Satellite AOT observations. **(a)** trajectory ending at 1500 m altitude does pass close to active fire objects but satellite AOT remains low. **(b)** trajectory ending at 3000 m altitude passes close to the fire objects followed by a sudden increase in observed AOT, indicating the source and age for the AERONET smoke observation.

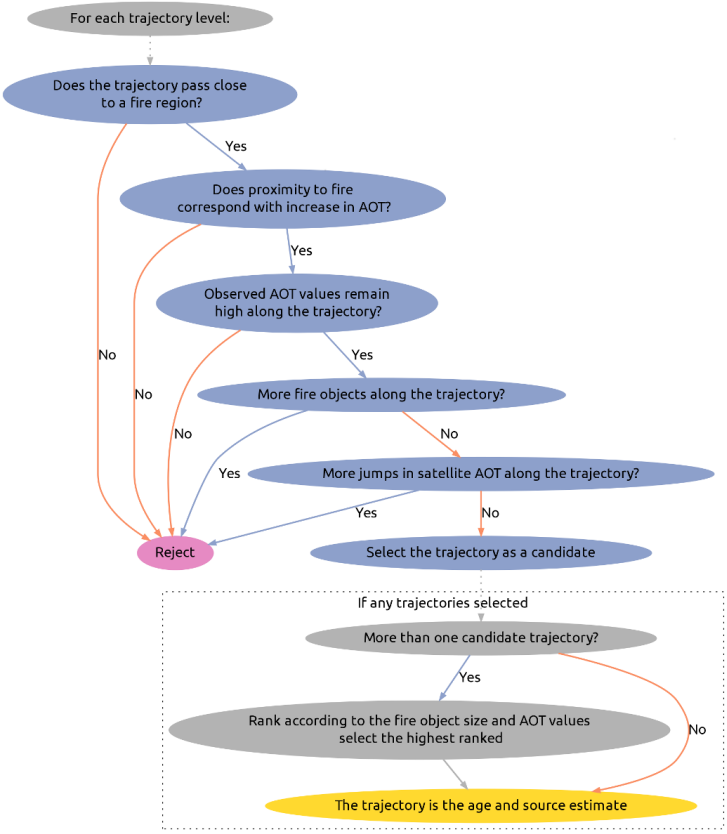

Figure 4. The decision tree used to identify potential smoke source and age. For every AERONET observation back trajectories arriving at different altitudes are tested against the conditions defined in the main part of the decision tree. Any trajectories identified as potential candidates are ranked (lower part of the diagram), identifying a single source and age estimate for the AERONET observation.

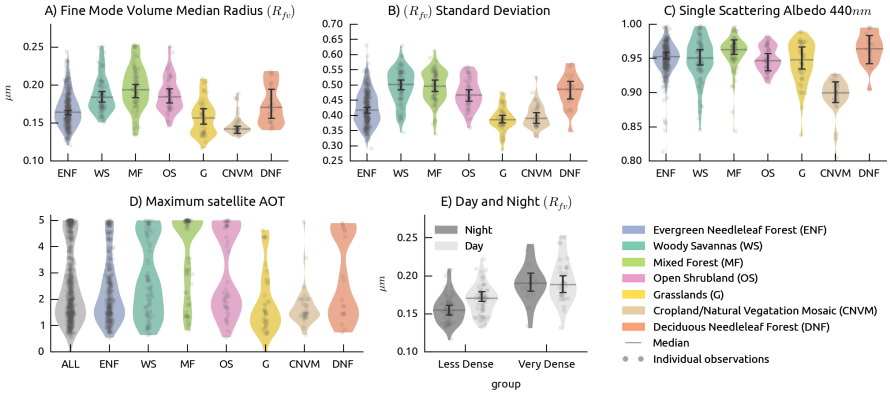

Figure 5. Optical and microphysical properties of smoke attributed to different land cover types **(a–c)**. Maximum satellite AOT detections along the identified trajectories **(d)** and fine mode volume radius distributions attributed to less dense and very dense plumes emitted during day and night hours **(e)**. Coloured areas indicate kernel density estimates, error bars represent 95% confidence intervals for median.

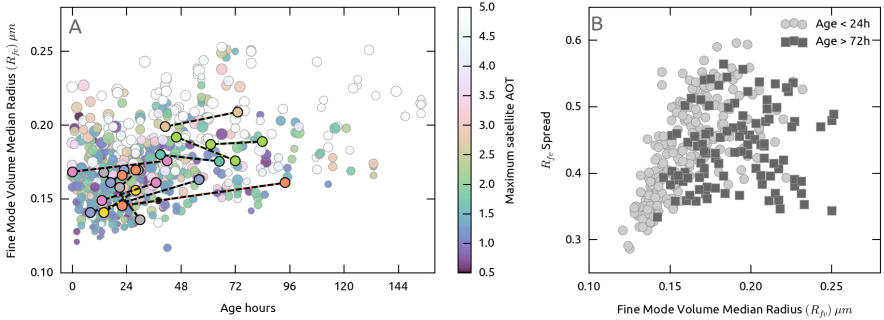

Figure 6. ~~Relationships between estimated age and AERONET retrieved plume properties.~~ **(a)** ~~fine~~ Fine mode volume median radius ~~,~~ **~~(b)~~** ~~Absorption Angstrom Exponent,~~ **~~(c)~~** ~~precipitable water content,~~ and **~~(d)~~** ~~fine~~ estimated age. Points joined by lines show identified paired observations. **(b)** Fine mode volume radius against ~~fine mode volume radius spread and smoke age estimates. Marker size in **(a–c)** indicates~~ the ~~SD from~~ spread of fine mode volume radius for young and well aged plumes.

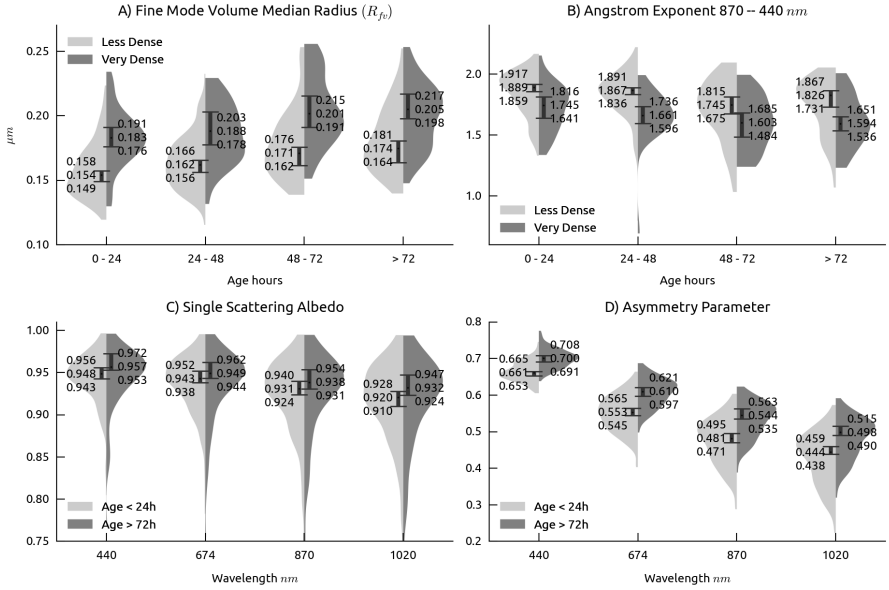

Figure 7. ~~Linear regression coefficients as change rates and initial smoke plume optical~~ Optical and microphysical properties ~~estimated for different land cover types **(a–d)** and for different land cover types~~ of young and ~~different wavelengths **(e–g)**~~aged plumes. ~~Shaded areas in top panels indicate 95CI~~ Less dense and very dense plumes binned per day of ~~all data fits~~ageing **(a–b)**, plumes under 24 hours old compared with smoke aged for more than 72 hours **(c–d)**. Error bars ~~representing~~ represent 95~~CI in **(e–g)** are shown only for 440nm~~ % confidence intervals for ~~clarity~~median. Numbers on the distributions represent high high CI, median value and low CI values.