Atmos. Chem. Phys. Discuss., 15, C2216–C2220, 2015
www.atmos-chem-phys-discuss.net/15/C2216/2015/

Atmospheric Chemistry and Physics Discussions

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

J. Reid (Referee)

reidj@nrlmry.navy.mil

Received and published: 4 May 2015

First, my apologies for the tardiness of review. For some reason I thought it was due in early May. This paper presents a system for evaluating the evolution of large smoke air masses by combining HYSPLIT lagrangian modeling with AERONET sun-sky retrievals. Hand analyses of along these liens have been done for years, but this work is novel in that they have hard wired a system together to allow for many more cases to be generated-which they then present. I think in general this is a nice bit of software

engineering, although I also think that analysis of the data is not so straight forward. Provided they mention and discuss all of the caveats that I list below, however I think it is suitable for publication in ACP.

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 $\sim$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.

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.

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. Thus, 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. 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. While I am gratified the authors point out their estimate of particle growth verified what I said years ago, in reality we are looking at different time scales. I would say their growth which is likely coagulation based is after the early fire dynamic time scales I looked at. A second form of sampling bias is that this work is only applicable to free troposphere plumes, and the authors should note that as impressive as the major plumes are in their coverage, there is considerable amount of nighttime or low smoldering smoke that does not make its way into the plume. So from a total burning emission budget point of view findings may be misleading. But, as long as one makes the caveat that only the large scale plume aspect of the fire is what is being monitored here, I have no problem.

Other minor issues I have is on source attribution. 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;). 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 get-

ting 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. 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. 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. All of the above discussion does not necessarily invalidate what they found. In fact given the communities proclivity to analyze major events, I think the system and findings are quite valuable on a case by case basis-especially in the verification of global models. But from a global biomass burning system point of view, I would be hesitant to draw conclusions.

Hope this helps, Jeffrey S. Reid US Naval Research Laboratory.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 6445, 2015.