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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Long-range transport of biomass burning smoke to Finland in 2006" *by* L. Riuttanen et al.

Anonymous Referee #1

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The manuscript describes some impacts of eastern European biomass burning on some ambient air monitoring stations located in Finland in the spring and summer of 2006. The paper follows the familiar model of monitoring and interpretive analysis. In my opinion, however, there is a difference between monitoring, monitoring with value-added analysis, and research. I believe ACP should be an outlet for the latter two categories. I didnot see how the paper produced anything fundamentally new, nor did it provide any long-term assessment for planning purposes. The paper in general reflected an inadequate knowledge of atmospheric chemistry and a weak literature search. For instance two recent major airborne campaigns in the boreal region (ARC-TAS and BORTAS) were overlooked. The English needs work. What is most troubling is that they show their analysis method (e.g. eqn 2) fails spectacularly on the NOx and CO lifetime and then use the method anyway, which is the reverse of the normal

scientific method. The paper should be rejected, but the data is likely of good quality and should be archived for possible future use in a different paper.

Specific comments

P4291, L17: There is a lot of data on boreal forest fires if that is what the authors really want. They have not done a literature search and only quote a single out of date biomass burning review that was never finished. More recent reviews of biomass burning EF are available (Akagi et al., 2011) and there were two major international airborne campaigns in the boreal region recently: ARCTAS and BORTAS (Palmer et al., 2013).

P4292, L6: After an extensive introductory discussion of boreal forest fires; here they say the fires were not boreal forest fires but crop residue fires. So what was the point of bringing up boreal forest fires?

P4393, L3-4: As shown in more detail below, the smoke age is not well-constrained and neither are the initial concentrations. If you don't know the age or the initial properties you cannot study the changes. That's just the way it is. On the other hand if the same smoke had passed over two SMEAR stations, then there could have been some useful analysis. E.g. changes in dO3/dCO between Varrio and Hyytial.

Top of page 4297: This whole section is disturbing. It seems impossible that the equation for the "fire sum" can be meaningful. What about when a back trajectory arrives at a location and it is not an overpass time or there is cloud cover so there is no information on whether something was burning at that moment? How is t-zero defined? What about trajectories that pass over multiple fires that can add new emissions? The fires will have different sizes and variable initial emissions. The uncertainty in smoke age is huge compared to the smoke age since the source region is much larger than the transport distance across the gulf. There will undoubtedly be some smoke of mixed age rather than a single (though un-measureable) age. Thus, the assumptions of the analysis are unrealistic so the analysis must fail and it does. The CO lifetime is about

twenty times too short and the NOx lifetime is about 20 times too long. The normal approach in science is to show that a new analysis gets the right answer for a known problem and then apply it to an unknown problem. Here it is shown that the analysis gets the wrong answer for multiple known problems. Normally when an analysis is shown to fail it is quietly discarded, rather than and published. To measure rates of processes one needs laboratory experiments that isolate the processes or Lagrangian measurements in single isolated plumes at the source and downwind as in Hobbs et al., 2003, Alvarado et al 2010, Akagi et al 2012.

P4297, L16-17: the concentrations will not decrease due to mixing when passing over a fire!

P4298, L1: Condensation > coagulation?

P4298, L3: The lifetime of the species discussed is already well-characterized via their known rate constants with e.g. OH. This is not a workable method to improve on that!

P4298, Eqn 2: the initial concentration, t, and tao are all unknown so the eqn is not useful.

4299, 7: Did the Hyy BTs always miss Hel? It is not specified and should be if any of the data is to be considered for publication on some other context.

4299, L17-19: It's not clear why data from years after the episode is being discussed when the pre- and post-episode values would be best and are shown in the figures.

4300: Re Table 2, dO3/dCO may be of interest. It seems that was not calculated though (see below).

4300. L23-24: The smoke impacts at Helsinki would have to be separated from changes in the local source strength that occur as e.g. high pressure builds and better traps locally-generated pollutants. NO and NOx are very short-lived so the large increases in these species during some smoke episodes strongly suggests an important local "co-contribution."

4302, L6: It's not just one study, but many studies that are better designed for determining NOx lifetimes that show NOx lifetimes that are much shorter than 63 h in smoke plumes. A 63h NOx lifetime would imply extremely low OH and extremely low OH would imply a much longer-than-normal CO lifetime rather than the much shorter one the authors also obtain.

4303, L5: The CO2 at ground level is strongly impacted by photosynthesis and respiration and the fire signal would be difficult at best to detect accurately at a distance. This is another unrealistic analysis.

4303, L13: One probably can't measure MCE at these low excess mixing ratios accurately that far away from a fire so no sense in discussing it. Section 3.1.3: the normal procedure would be to compute dO3/dCO from (O3smoke - O3background)/(COsmoke - CObackground) – not sure what the authors are doing here?

4304, L20: The O3 production depends on NOx/CO at source, but one has to measure NOy/CO downwind to estimate what NOx/CO was originally.

4305, L3: Wrong speculation: NOx tends to increase O3 prod in BB plumes not decrease it. Section 3.1.7. In general the best indicators of fire are acetonitrile, HCN and others. None of which were measured in this study. The species measured here all have multiple sources and are not well suited for quantitative source apportionment.

4307, 23-25: I'm not a particle expert, but this seemed potentially interesting as the possible topic for a different shorter paper "Fig. 4h, with high nucleation mode concentrations and low accumulation mode concentrations for clear sky conditions and vice versa for smoke episodes." But then this observation appears to be contradicted later on page 4311 (see below)?

Section 3.2. This whole section is invalid.

P4311, 13-16: The authors seem to say the nucleation mode could be transported from closer fires, but it could also be from NPF involving upwind fires followed by growth, not

sure how you could tell the difference. In any case, regarding growth, wouldn't the nucleation mode at 20 nm have insignificant mass compared to the similar amount of particles at 200 nm in the accumulation mode? How is this consistent with the earlier plot in Fig 4h where the nucleation mode was suppressed in smoke events?

4312, L2: coagulation conserves mass.

P4312: At this point, the paper wanders in an unfocused manner.

Conclusions: I don't agree that the new term "fire sum" is meaningful and it is also not explained well.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 4289, 2013.