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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Aircraft-measured indirect cloud effects from biomass burning smoke in the Arctic and subarctic" *by* L. M. Zamora et al.

Anonymous Referee #1

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General comments: The authors analyze datasets from a number of studies to examine the influence of biomass burning (BB) particles on Arctic clouds. It is a difficult undertaking, since there are not only many datasets but also many different instruments. The subject is important, for the reasons the authors discuss, and I think the authors have done a good job of estimating some potential effects of BB particles on Arctic clouds as the title describes. That said, there are improvements needed before the paper is worthy of publication in ACP. The estimates of radiative forcing need to be clarified, as does the use of the term "background", and there is a lot of speculation made in Section 3.3 that is not substantiated by the observations and adds considerably to the

length of the paper. Detailed comments follow.

Major comments:

1) The radiation forcing estimate given in the abstract, discussed on page 22844 and again in the conclusions needs clarification. On Page 22844, you say "Therefore, the -2 to -4 W m-2 range is only applicable in the subarctic in some conditions. Nonetheless, this estimate at least provides a rough indication of how important these effects might be." Putting aside the surface albedo, is the -2 to -4 W/m2 estimate for local effects by BB on clouds, or is it based on some anticipated coverage of the Arctic by clouds and BB plumes? Also, most of the observations were from studies conducted during springtime. Is your forcing calculated for the spring or does it include the summer too when the sun is higher and the albedo is lower? Please elaborate.

2) Section 3.3 uses four pages and five figures to suggest that coagulation of particles associated with a clean environment might influence the hygroscopicity of BB particles by up to 10-20%. It relies on one reference (Lohmann and Leck, 2005) and later adds a second (in section 4; Lawler et al) to suggest the hygroscopicity of smaller particles in the Arctic may be relatively high. This process may be worthy of mention, but there are many things discussed in this section that are speculative without sufficient justification; I have made several specific comments about this below. The presentation related to this section needs work, and overall I feel it detracts from the main aspect of the paper already presented. This section really forms the basis for a different paper, and I think it should be treated that way or the presentation should be made much more concise.

3) Use of the term "background". Page 22833, lines 17-21 – These concentrations are high, particularly the sulphate and BC values. They are not "background" values. The sulphate and BC values (<0.9 and <0.3) represent Arctic Haze. They are reference values for your BB assessment, but the use of the term 'background' is inappropriate. Page 22834, lines 1-7 – The CO levels mentioned here are clearly not background values. They too are simply reference values for BB. Values of 0.2 ppb of acetonitrile

can be found over the ocean (e.g. de Gouw et al., J. Geophys. Res., 108, 2003). On page 22835, line 3, you put background in parentheses, whereas everywhere else it is not. Replacing background, everywhere used, with "reference" would suffice. Additonal related comment - You appear to be mostly considering direct hits of the BB plume on the cloud. But BB plumes may disperse and dilute leaving lower concentrations of BB particles available to still influence cloud, and such influence could be relatively more significant in the long run (e.g. less impaired by competition for water vapour).

4) Page 22830-22831, first two paragraphs of section 2.2.2 – There is very little about the qualities of either the CAPS-CAS and the FSSP-100 droplet measurements. The FSSP-100 measurements are at least discussed relative to other independent measurements (LWC from hot-wire), but it seems that the CAPS-CAS observations are assumed to be of high quality without any demonstration of such. Based on the LWC and N(liq) in Table 8, the mean size of the volume weighted distribution varies between about 5 um diameter to 3.5 um diameter, which means that about half of the LWC and most of the droplet numbers are below those diameters. How accurate was the CAPS-CAS in 2001, when the measurements were made, at measuring droplets below 5 um diameter? What are the consequences if those measurements are of relatively poor accuracy?

Minor comments:

5) Page 22831 - A comment on potential artifacts from droplet shattering on the probe tips (e.g. Korolev et al., B. Am. Meteorol. Soc., 92, 967–973, 2011.2011). The reference is for ice crystals, but very large droplets may also shatter creating artifact droplets. It is likely a non-issue for the mostly smaller droplets you measure, but could be important for some of the reference measurements.

6) Page 22832, lines 19-22 - Understandable, but the horizontal extent of a cloud and the number of times it will be sampled by an aircraft may be related: it is a tendency in these studies to sample clouds of greater horizontal extent more than smaller clouds.

Since larger clouds will have a greater radiative impact, should they not be considered more than smaller clouds? It might be different consideration if you were examining a process only, but you are considering an impact here. Does your approach potentially bias the impact lower?

7) Page 22833, lines 8-15 - Do the LWCs relate more to Re or N(liq), which may tell you something about the mixing processes?

8) Page 28334, line 8 – Here, do you mean high-quality or high-resolution?

9) Page 22835, line 15 – In the literature, there tends to be a generic use of the term Aerosol-Cloud Interactions that pervades the indirect effect. Are you not just assessing the effect of the BB aerosol on cloud? Is there an interactive aspect implicit in what you are assessing here? You do not deal with deposition resulting from precipitation altered by the aerosol in a meaningful way, other than to mention it at the bottom of page 22849. A few words of clarification would be helpful.

10) Page 22835, on line 26, you refer to CCN, which is not defined anywhere previously, including the abstract where it is mentioned as CCN. Please define it in the abstract. On line 28, background values of 0.018 are referred to as being subtracted. What are the units and are you referring to CO or CH3CN or something else?

11) Page 22836, lines 21-24 - Both the UHSAS and the APS use sheath air to focus the particles for detection. The sheath air is normally dried and that can also help with the drying of the particles prior to detection.

12) Page 22837, lines 5-6 – Would you please clarify how this uncertainty can be "fully eliminated in model simulations"? It reads to me as if we don't need observations, since the model can solve the problem.

13) Page 22837, line 15 and 17 – insert "e.g." in front of these references, here and elsewhere (22840). The competition process was demonstrated 30 years ago.

14) Page 22838, lines 10-11 – It seems odd that there were no inversions topping the

clouds. Even in the typically stable environment of the Arctic, the layers will be defined by slight inversions. How were they contained?

15) Page 22838, line 19 – It is surprising to see CO up to 500 ppbv classified as out-of-plume, when the previous discussion referred to much lower values of CO as the reference for non-BB. What was the basis for identifying the plume?

16) Page 22843, line 6 – Is ice "typically well mixed throughout" during the summer?

17) Page 22844, line 22 – coagulation is usually a term associated with aerosol particles, whereas cloud processes refer to collision-coalescence.

18) Page 22848-22849 – Can you briefly discuss how does CCN number vs CCN hygroscopicity plays into the impact of BB on the ACI index?

19) Page 22850, lines 20-21 – sulphates are not necessarily "an additional organic component".

20) Page 22851, line 1 – change "condensation of external particles onto" to "coagulation of external particles with".

Specific comments related to Section 3.3

21) Page 22845, lines 11-13 – Please add to your references: Leaitch et al., Elementa, 2013 and Tunved et al., ACP, 2013.

22) Page 22845, lines 16-20 – During the time of Arctic Haze influence, 1) there are generally few particles smaller than about 80 nm, and 2) the presence of the larger particles inhibits the formation of smaller particles. So when the aerosol is dominated by Arctic Haze or BB influences, the "small background aerosols" are not directly significant for liquid cloud formation. However, during the summer, the air is quite clean and there is potential for such small particles to be important for clouds (e.g. Leaitch et al., Elementa, 2013). Please do not generalize here.

23) Page 22846, line 3 – Why do you use backscatter here instead of total volumetric

scatter? The relative backscatter is higher for smaller particles, but their total scatter is generally smaller reducing sensitivity to them. What is the detection limit for the backscatter observations?

24) Page 22847 – The discussion of the rapid change in CN is hampered by 1) the absence of a discussion of the possibility of new particle formation (NPF) aided by a sharp reduction in the condensation sink (as indicated by the APS and OA; the backscatter observations appear to have a delayed response relative to the OA), 2) the failure to plot the data as vertical profiles rather than time series. It is difficult to understand from the time series the regions of mixing/transition region(s) in which the coagulation is apparently taking place. If you must retain this discussion, please make it easier for the reader by plotting the data as vertical profiles. The explanations that "Such a rapid change in CN(TSI) concentrations could be explained by either a sharp non-mixing transition zone or by rapid coagulation of the small particles onto the larger haze particles" seems to avoid the possibility that NPF associated with a small condensation sink may explain the rapid increase in CN. Certainly small particles will coagulate with larger particles if present together, but it seems that these layers are relatively de-coupled and that the higher CN concentrations after 69500 are more likely to be the result of NPF in very clean air.

25) Page 22848, lines 5-21 - Were there any CCN measurements of the BB particles that would suggest larger hygroscopicities (kappa values) than expected for a "pure" BB aerosol, exclusive of sulphate? How important an influence on the hygroscopicity would this coagulation be relative to the smaller amounts of sulphate found in the BB particles? You mention sulphate in Section 4, but not here.

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