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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Sources of light-absorbing aerosol in arctic snow and their seasonal variation" *by* D. A. Hegg et al.

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I focus on the analytical technique and receptor modeling, since these form the basis of the results and discussions that follow.

Methodology:

Since the Grenfell et al. (2010) paper is reported as "submitted to Applied Optics", the methodology description here is inadequate. It is not quite clear how a distinction is made between BC and non-BC LAA, using just a "maximum BC concentration" and the assumed Angstrom absorption exponents. There are two unknowns (BC and non-BC LAA), and one equation; the other is an inequality. It appears there could be numerous solutions to such a problem.

Further, since the Grenfell paper is under review elsewhere (in a non-open access journal), the authors should explain why they choose Angstrom Absorption Exponents of 5.0 for non-BC LAA. Earlier work by Kirchstetter et al. (2004?) suggests an AAE closer to 2.0. Also, how valid is the assumption that the AAE of non-BC LAA remains constant? Clearly, as the authors point out, the absolute magnitude of the two LAA components is not as important as the trends for PMF analysis. But if the absorption cross-sections and AAE vary depending on the combustion conditions and/or both values are assumed incorrectly (6.5 m2/g and 5.0 for non-BC LAA both seem questionable), then will that not affect the PMF results adversely?

PMF:

There appears to be an assumption by the authors that the major source of BC in the Arctic is biomass burning, based on the Hegg et al. 2009 study, and that BrC is also largely derived from biomass burning. However, as the authors themselves report, at least numerical models suggest that fossil fuel combustion is the major source of BC in the Arctic! Since the manuscript under consideration tries to determine whether the sources of BC and non-BC-LAA are the same, this assumption seems dangerous. In my experience, long-range transport tends to co-mingle different sources, leading to erroneous PMF results; thus, making such an assumption seems counter-productive.

There appears to be a disconnect between the measured, direct source profiles by Hays et al. and Oros/Simoneit, and the PMF source "profiles." The former suggest vanillin/levoglucosan ratios orders of magnitude lower than the PMF results as described on page 13764. Why do the authors see so much more vanillin relative to levoglucosan? One alternative explanation suggested by these PMF source "profiles" could be that what the authors claim to be crop/grass burning with a vanillin/levoglucosan ratio of 0.5 is actually from boreal biomass burning (which is reported by Oros/Simoneit to be 0.1-0.3). The PMF source with vanillin/levoglucosan ration of 80 could be something altogether different!

On page 13766, the authors claim it is unlikely that soil dust can be "so highly correlated" with biomass burning that it cannot be resolved from the biomass burning source. This speaks to my doubts about PMF's performance with emissions transported over long range. Further, when a forest or grass burns, surely there is some soil dust that gets resuspended (or even gets burned) in the smoke. So I would not be surprised if soil dust-sourced Fe contributes to some of the non-BC-LAA. The authors' second explanation, that not all of the non-BC-LAA can be accounted for by Fe, seems more plausible. (At the same time, look up my comment about the authors' method in discriminating between BC and non-BC-LAA – they could be under- or over-estimating BrC and BC.)

Finally, I am also skeptical of the use of PMF to distinguish between combustion sources. I note that the authors have not used *any* organic markers for fossil fuel combustion, like hopanes or steranes, even 15 years after Schauer et al. published their seminal work on CMB with molecular markers. Admittedly, PAHs may not be much help, but neither are metals. Given the alternative explanation from numerical models that fossil fuel combustion might be a major source of LAA to the Arctic snow, I find this neglect, in a sense, to be a glaring error in this study.

Before addressing the discussion and conclusions, I would like the authors to respond to these critical issues.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 13755, 2010.