Atmos. Chem. Phys. Discuss., 12, C10743–C10746, 2012
www.atmos-chem-phys-discuss.net/12/C10743/2012/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Temperature-dependent accumulation mode particle and cloud nuclei concentrations from biogenic sources during WACS 2010" *by* L. Ahlm et al.

Anonymous Referee #2

Received and published: 19 December 2012

The authors report simultaneous measurements of organic aerosol source classes and their potential influence on CCN concentrations using a suite of instruments at two different sites in Whistler, Canada, during the WACS 2010 campaign. The significance of the work and presentation of the material are suitable for publication in ACP. While the general conclusions seem reasonable, a few details should be addressed before publication.

Comments:

It is not clear that the four periods categorized by Macdonald et at. (2012) are definitive for discerning influences of various sources to the site—which seems to be one of the

main objective of this manuscript—and a separate set of indicators for source influence (e.g., temperature) are described in various places throughout the text. For instance, Periods 2 and 4 are very similar with respect to the aerosol/gas-phase measurements, while Period 2 is actually comprises two periods relevant for chemical interpretation (anthropogenically dominated vs biogenically dominated). As such, this manuscript would be strengthened by a contingency table showing the relationship between the periods from Macdonald et al. (2012) and periods outlined in the first two bullet points in Section 4. There are statements in the manuscript to the effect that Period 4 has some biomass burning events, but is not borne out by many of the analysis presented in the manuscript (e.g., the PMF analysis on FTIR spectra or STXM/NEXAFS analysis), so it seems this complexity can be neglected (to a first order) in presenting this information.

Regarding correlations of PMF factors with particle number concentrations, one interpretation which is not stated explicitly is that during biogenic-aerosol dominated periods, most of the accumulation-mode organic particles are biogenic, whereas this is not necessarily the case during combustion source-influenced periods (presumably because there is OM from biogenic sources mixed in).

I can appreciate that the authors derive the source class based on correlations with XRF elemental composition and other measurements without consideration for spectroscopic signature, but the significant departure in factor profiles from previous assignments for similar source classes (Schwartz et al., 2010; Takahama et al., 2011; Russell et al., 2011) should be noted. Even the variability in quantified functional group abundance between the two sites is significantly different, especially with respect to the carbonyl and carboxylic acid fractions (Factors C and D) (the profiles look quite similar but can this quantification be correct?). This seems to be inconsistent with the conclusions of Russell et al. (2011) regarding broad similarities in composition among PMF profiles attributed to common sources classes. Why was FPEAK=0 chosen in both cases (Figure 3)? Is there another interpretation of the factor analytic decomposition that would make more sense? It is not clear if these correspond to the minimum

value of the objective function, but as discussed by Paatero et al. (2002), the rotations performed by PMF are approximate and some amount of increase in Q-value is acceptable among alternate solutions generated by the algorithm.

p. 28004, lines 16-18: The wording is not entirely correct in interpreting single-particle measurements. The alkene/aromatic groups were not "attributed [...] to the influence of carbon tarballs," but they indicated that the selected particles examined were tarballs observed in biomass burning episodes, as reported previously by Tivanski et al. (2007).

p.28008, second paragraph: The authors draw upon correlations between number concentrations of particles less than 100 nm and a particular PMF factor to support their assignment of this factor to a predominantly combustion source. As these nucleation-mode particles (likely to be largely BC in composition) are probably not the particles measured by FTIR, the authors should include available citations to studies showing the mass size distribution of OM from combustion sources in the accumulation mode—which is what is presumably measured by their FTIR—to make this argument more convincing.

There is some mention of periods during which the WRN and WHI measurements are decoupled because of boundary layer meteorology. Would not the manuscript provide additional contributions to the community by focusing on differences in aerosol composition below and above the boundary layer, if such distinction can be made from archived meteorology (e.g., HYSPLIT), remote sensing information (McKendry et al., 2011), and the suite of in-situ instruments at the sites?

Figure 6: The top spectra seems to include some saturated spectra.

Figure 8: Would it make sense to segregate the profiles based on either the periods 1-4, or combustion and biogenic source-dominated periods? It is difficult to assess the relative contributions from the different source classes otherwise (e.g., is this diurnal profile in BC at the site observed during biogenically-dominated periods).

Figure 10: What is implied by the linear model (as indicated by regression lines) between number concentration and temperature? Related to a comment by the other reviewer, as correlation coefficients indicate the degree of linear relationship, they are not often the ideal metrics to use in environmental analysis–though indeed they are widely applied out of convenience. However, correlations can be useful to examine relationships provided that the degree of nonlinearity is small, or the dynamic range is small enough that the functional relationship can be approximated with a linear one. But fitting a linear model suggests a stronger interpretation—what physical significance do the slopes and intercepts have?.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 27989, 2012.