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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Long-term volatility measurements of submicron atmospheric aerosol in Hyytiälä, Finland" *by* S. A. K. Häkkinen et al.

Anonymous Referee #1

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The manuscript describes measurements of aerosol volatility in a remote Boreal forest. The measurements are unique in that they continuously span a period of over two years, whereas many other field studies are of much shorter duration. Overall the data are well presented and the analysis is sound. However, the authors need to address several issues in the discussion and interpretation of the results before the manuscript will be suitable for publication.

(1) The manuscript makes two conclusions that are never related to one another or resolved. The first is that MFR correlates with PAH concentrations, and thus anthropogenic activity. The second is that MFR correlates with organic nitrates, which I assume must be secondary in nature. How are these two phenomena correlated? Is MFR highest when air masses pass over populated areas (anthropogenic influence)

and then have sufficient time (12+ daylight hours) for photochemical processing? This could be inferred from the back-trajectories. One could postulate that the lower-volatility PAHs are picked up in urban areas, stay in the condensed phase because of low volatility, and are joined by anthropogenic nitrate-containing SOA that is formed when NOx is high. It is critical to connect these two conclusions, or at the very least note that they are seemingly at odds, rather than to leave them as mere observations.

(2) One important result seems to be the presence of low-volatiliity OA that potentially contains organic nitrates. This message is buried in the manuscript after much minute detail about what some may consider less-important topics. Authors should consider moving this info to a more prominent location in the manuscript.

(3) The concluions section should be more focused, not merely a reiteration of earlier portions of the paper. Much of the first few paragraphs of the conclusion are repetitve from earlier in the manuscript.

(4) Page 11206-11207 The meaning of DMPS should be introduced with the instrument in section 2.2.1, not as a side note about VDMPS in section 2.2.2.

(5) Page 11207 TD residence time was "around 1 second." It would be preferable to know the centerline residence time at ambient temperature

(6) Section 2.2.5 - Please add PAH filter sampling to Figure 1.

(7) Were the DMPS and VDMPS compared at ambient temperature? It seems that the two instruments are different, with different size ranges (20-500nm for VDMPS, 15-1000nm for DMPS). How was this size range discrepancy resolved?

(8) Section 3.2 - what fraction of the BC mass is expected to be in particles 500-2500nm? It seems that most of the mass would be in that size fraction. Quantify the effect of this estimate on MFR_non-BC

(9) Page 11214, Lines 5-6 "During winter and fall periods, the temporal variation of the MFR values was relatively similar independent on the TD temperature." What exactly

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does this mean? Please clarify.

(10) Page 11214, Lines 6-8. "During the spring and summer months, however, the MFRs below 200 âŮęC seemed to show an increasing trend while the MFRs at higher temperatures were decreasing." I never see the "low temperature" MFR moving a different direction than the "high temperature" MFR. In the fall/winter the higher temperature MFR level off while the lowtemperature MFR slightly increase. Also from winter into spring, the higher MFR seem to have a bigger downward slope than the lower temperatures.

(11) Figure 4 - are these raw MFR or MFR_non-BC?

(12) Page 11214, Line 20. "Residence times and aerosol type varied from study to study" This is important, because residence time matters in the TD. Make this point more strongly.

(13) Table 1 can probably go into the supporting info. I don't think it's critical to interpreting the TD results.

(14) Figure 5: it's hard to distinguish between black and grey points.

(15) Page 11215, Lines 21-26 - Does the calculation following Riipinen's method include a BC fraction? Or is this strictly for the measured MFR represented by a single-component organic particle?

(16) The caption to Figure 5 states that the line assumes that "all nonvolatile aerosol mass is BC". This implies that some of the OA is "nonvolatile". I disagree with this in an absolute sense. ALL OA is semivolatile. It is apparently nonvolatile in the context of your TD, residence time, etc.

(17) I recommend against circling the r^2 in figure 6.

(18) Page 11216, Lines 12-14 "although speciïňĄcally in summer the black carbon concentrations were often of similar magnitude than the concentrations of the non-volatile

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material. " Again there is confusing interchange between BC and nonvolatile material. BC is truly nonvolatile. I suggest changing the wording and calling the non-evaporating, non-BC fraction of the aerosol something else. Perhaps "non-evaporating".

(19) Page 11216, last line. "non-volatile particle mass fraction" requires some thought to deconvolute. It probably gets boring to keep writing MFR, but it is easier for the reader to interpret. Also, it implies that everything that does not evaporate in the TD is nonvolatile.

(20) Page 11218, lines 3-4 "It is possible that some of the PAHs having low saturation vapor pressures, around $10-7$Pa." The mass fractions of the PAH (tot PAH always < 1%) are not sufficient to drive significant differences in MFR. Also, some of these PAH have C* > 1 ug/m3. The PAH seem useful as an indicator of anthropogenic influence, but are not a major component of OA or the MFR.

(21) Page 11218, middle paragraph - I suggest moving this up, immediately after the other paragraph discussing ambient temperature.

(22) In general section 4.4 could benefit from some reorganization. There is a sometimes confusing mixture of variables (temperature, trace gases, etc) and seasonality. I suggest the authors try to me more systematic in the discussion. I.e., focus on seasons first, then variables.

(23) Perhaps switch Figures 7 and 8. Figure 7 is introduced first, but only briefly. The major discussion of it is after the major discussion of Figure 8.

(24) Figure 10 is not discussed enough to make it essential to the manuscript. I recommend moving this to supporting information. In general, the last paragraph of section 4.5 seems superfluous and/or misplaced. It seems to show redundant info as Figure 9 - high correlation of aerosol nitrate with MFR_non-BC

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 11201, 2012.