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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Hygroscopic properties and mixing state of aerosol measured at the high altitude site Puy de Dôme (1465 m a.s.l.), France" *by* H. Holmgren et al.

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We thank reviewer 1 for his helpful comments and suggestions. We have addressed the comments point by point below.

Minor comments:

Comment 1: Page 6769: If local home heating devices contribute to aerosol with very low GF values (1.0-1.1) when the site is influenced by the PBL in winter, then what fuel is likely to be used? Biomass burning aerosol is associated with much higher GF values earlier in the text, presumably if the inorganic aerosol content is high, or

if secondary inorganic ions are internally mixed. The very low GF values observed here point to either BC-rich biomass burning aerosol or BC-rich particles associated with combustion of different fuel. Some discussion of potential heating fuels would be useful here. Referring to previous SP2 measurements of fossil fuel and biomass burning plume BC mode diameters may also be useful, for example (Schwarz et al. 2008). Furthermore, if the seasonality controls the PBL intrusions, isn't it possible that more fossil fuel combustion aerosol, associated with traffic, for example, is observed in the winter because of the PBL effect.

Reply 1: On page 6766, lines 16-18, we state that BC and mineral dust has a GF of less than 1.05, while biomass burning shows a large range of GF that can go up to 1.65. However, these high values were measured after the biomass burning particles have had some time to mix with secondary inorganic ions, and much smaller GF were measured closer to the source. This has been clarified in the text. According to the referee comment, it has also been added that the very low GF values observed in this work point to BC-rich biomass burning aerosol or BC-rich particles associated with other combustion sources, such as fossil fuel combustion, which have had little time to mix with secondary inorganic ions. Moreover, we clarified the role of the seasonality of PBL intrusions on the seasonality of the hygroscopic behaviour of particles (see answer to reviewer 2's comment no. 9).

Comment 2: Page 6770, first paragraph: Particles with relatively high GF values may still contain BC. If the inorganic ion volume fraction is very high, it is not possible to assess whether a small BC core is or is not present using this method.

Reply 2: Yes, this is true. We have now mentioned this in the text.

Comment 3: Page 6773: Is it possible that partitioning of nitrate or ammonium nitrate at lower temperatures at night also contributes to the higher night time mean GF values?

Reply 3: Yes, it is indeed possible and we now mention this possibility in the text.
Comment 4: Page 6775, line 1: Do the authors mean that the primary marine particles

are internally mixed? Displacement of sea salt chloride by nitric acid during mixing with anthropogenic plumes? Or do sea salt particles have a minimal contribution to number at 165 nm? A GF of 1.8 would be too high for an OA-ammonium sulphate internal mixture, but it may be reasonable for a particle composed predominantly of sodium nitrate.

Reply 4: A GF of 1.8 would indeed be reasonable for aged sea salt particles, either by displacement of sea salt chloride by nitric acid, or by mixing of NaCl with less hygroscopic species such as organic, ammonium sulphate or ammonium nitrate. We now precise these possibilities in the text.

Comment 5: Fig. 10 caption: How is this seasonal variation? Is this not classified based on air mass origin?

Reply 5: This is a correct observation; the caption is wrong and has been changed.

Comment 6: Fig. 11 is perhaps not necessary as the GF depends more on season than air mass origin as discussed in the text.

Reply 6: We believe that Figure 11 is necessary, as we show by splitting seasonal variations by air mass types, that the general seasonal variation (without air mass splitting) is not observed in individual air mass types. In fact both the seasons and air mass type influence the hygroscopic properties of the aerosols at puy de Dôme.

Specific comments:

Comment 7: Page 6765: Should be “Matlab”

Reply 7: Lines 10, 16 and 18 - Yes, this has been changed.

Comment 8: Page 6766: should be “biomass burning aerosol”

Reply 8: Line 18 – Yes, this has been changed.

Comment 9: Page 6766: Duplissy et el. 2011 observed higher GF values for aged

SOA in simulation chamber studies (Duplissy et al., 2011)

Reply 9: Line 21 –This information, and the reference, have been added.

Comment 10: Perhaps shorten the discussion around Fig. 11. The main point is that there is a dependence upon season but not necessarily air mass origin.

Reply 10: see previous reply to comment 6.

References: Duplissy, J., DeCarlo, P. F., Dommen, J., Alfarra, M. R., Metzger, A., Barmpadimos, I., Prevot, A. S. H., Weingartner, E., Tritscher, T., Gysel, M., Aiken, A. C., Jimenez, J. L., Canagaratna, M. R., Worsnop, D. R., Collins, D. R., Tomlinson, J., and Baltensperger, U.: Relating hygroscopicity and composition of organic aerosol particulate matter, Atmos. Chem. Phys., 11. 1155-1165, doi:10.5194/acp-11-1155-2011, 2011.

Schwarz, J. P., Gao, R. S., Spackman, J. R., Watts, L. A., Thomson, D. S., Fahey, D. W., Ryerson, T. B., Peischl, J., Holloway, J. S., Trainer, M., Frost, G. J., Baynard, T., Lack, D. A., de Gouw, J. A., Warneke, C., Del Negro, A.: Measurement of the mixing state, mass, and optical size of individual black carbon in urban and biomass burning emissions, Geophys. Res. Lett., 35(13), L12810, 2008.

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