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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol cloud activation in summer and winter at puy-de-Dôme high altitude site in France" *by* E. Asmi et al.

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We thank Referee for their nice words, and we are very grateful for the comments and remarks, which greatly helped us to improve the manuscript. Our answers are specified below. We largely agreed with the suggestions, and have done our best to improve the manuscript accordingly.

Comment: Page 23046, line 20: Could the authors explain in more detail how these new errors were derived?

Reply: We made sensitivity studies using van't Hoff factor of $\pm$ 0.2 around the preset values and $\pm$ 5 K deviation of temperature which led to standard deviation of SS of roughly 0.02 % and 0.01 %, respectively. In addition, due to a higher quantity of doubly

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changed particles at larger mobility (smaller size and higher SS) region (as a result of the shape of the atomized distribution), we estimated an additional 0.01 % deviation to follow from this. This is explained in the revised manuscript. We find these error estimates, where additional uncertainties are taken into account, more realistic than those defined purely from different calibrations.

Comment: Section 2.1.3, 1st paragraph: Is it possible to quantify this error – what sizes are significantly affected, at what wind speeds, and by how much are the concentrations / size distributions underestimated?

Reply: The inlet cut-off diameters are calculated using the flow rate (112 m3/h), diameter (94 mm) and mean wind velocity (6 m/s). And yes, it is possible to theoretically evaluate the losses of droplets at higher wind speeds, but not the impact on losses of corresponding residual particles, as we do not know the correspondence between initial droplet size and residual particles. We find this an important issue, and more extensive experimental tests and comparison with theoretical values have been planned.

Comment: Page 23052, lines 14 – 16: The authors seem to be talking about a "persistent air mass" during the first part of the summer experiment, but from figure 2, it seems this period of increasing organic fraction is characterised by three different air masses. Could the authors please clarify this?

Reply: Looking at the figure 2 this is true. Even though aerosol chemistry showed some defined patterns with air mass origin, at this stage of the manuscript it is too early to discuss this. Connection of air masses to aerosol chemistry and organic mass fraction becomes better visible e.g. in figure 9. Only from figure 2 it seems a higher temperature, possibly indicating more southerly air flow and increased biogenic activities, typically correlates with increased organic mass fraction. Text was shortened and modified to match the figure.

Comment: Page 23054, lines 1 – 4: In this statement, the authors are not taking into account possible seasonal variation. How do the CCN to CN ratios compare between

ACPD

12, C9531–C9535, 2012

summer and winter in the same air mass (marine or continental). There seems to be enough data to be able to answer this question in a quantifiable way, and the authors could use this to further justify their statement.

Reply: We agree, and in fact this is studied in connection with figure 7. Therefore, also this chapter and discussion on p. 23054 was moved forward and discussed in chapter 3.3 “Aerosol CCN properties by air masses”.

Comment: Page 23054, lines 6 – 7: The size distributions on their own don’t fully explain this – composition also has a role as the authors themselves stated in the previous paragraph. Please clarify this statement.

Reply: Yes, this was too strongly written as so, since size distributions are only a partial explanation. This part of the text was also modified and moved to chapter 3.3. We also studied the connections of aerosol kappa and size distributions more carefully and modified the figure 5 accordingly. In the revised version of the manuscript we conclude that in our study case, the summer variability of CCN to CN ratios was more explained by the variability of aerosol kappa than that in winter, which reflected largely changes in aerosol size distributions.

Comment: Page 23054, lines 25 – 28: The authors say that the summer average kappa “fits well to suggested global average continental aerosol kappa of around 0.3” after stating on the same page (see above) that measurements in summer “represent mainly marine aerosol”. Again, please clarify.

Reply: Very good point. While the measurement site was about 400 km from the nearest coast, the aerosol had always a strong continental influence. Truly, kappa values of around 0.7 are typically measured only at coastal regions and over the oceans. At European continental sites, the kappa values are more “continental” even at air masses of marine influence. We clarified here that “continental kappa” is used to refer to kappa values measured at continental sites.

ACPD

12, C9531–C9535, 2012

Comment: Page 23055, line 13: Please explain the choice of grid size and why they are different between summer and winter.

Reply: Yes, this was only due to the fact that in winter the air masses came from wider longitudal area. Selecting different grid sizes did not affect interpretation of results. However, picture looked better when not too many "NaN"s were in between the grid squares. We added a sentence: "In summer the air masses came was from smaller geographical area which enabled us to use a better resolution grid size with approximately the same statistics per square."

Comment: Page 23055, lines 14 – 20: Please explain the choice of weighting in more detail. Are these weightings realistic?

Reply: It is not straightforward to define the "impact" of a region (or a square) to a measurement made in a single location. Due to removal processes the near-by squares have a greater impact than squares at further distances. But this does not vary greatly between different trajectories due to relatively constant transport times which make this more of a constant distance dependent factor without too large influence on the results. Instead, the particle and vapour sources are in emitted mass or number per time unit. Therefore, the longer the air mass spends in a certain square the greater will be its impact on the measured concentration. Here we find this simple linear approximation is well justified, and makes also an impact on the results. Based on these, we decided to use the time and temporal distance in the weighting to get an idea of importance of different regions on measured aerosol quantities. Of course, in reality, the losses are not constant in time as assumed, and depend also on which aerosol quantity is in question (e.g. mass or number). However, without using an aerosol transport model we find this is a realistic attempt to approximate regional impacts and relative differences. Comparing it with an approach where no weighting is used instead, it emphasizes the results obtained when a certain square actually had a possibility (time) to make an impact for the measured result. By no means, however, we consider it representative in comparison to modeled results but do consider it more representative (on average)

ACPD

12, C9531–C9535, 2012

than an approach without weighting.

Comment: Page 23055, lines 15 – 19: This is a very good point and deserves a little more exploration. In particular how does it affect the conclusions on CCN to CN ratios (top of pg23054), kappa (section 3.3) and organic fractions in marine air? It would be useful to have a paragraph with a general discussion on the effect of the mountain on mixing of air masses along with specific mention of the issue when discussing characteristics of marine air mass. The limitations of back trajectory analysis should be noted.

Reply: Here we are not sure of which point the referee is talking about, since p. 23055 lines 15-19 discuss on trajectory weighting. Could we get some more specifications on this question?

Comment: Page 23061, lines 12 – 14: It is no surprise that removing outliers improves a fit, but you cannot simply remove points just to improve a fit. You need to be able to justify removing these points. Unless the authors can explain why they are outliers and why they can be removed, I would recommend removing this sentence.

Reply: This is true. Data from summer seems to be more scattered (possibly also due to lower time resolution). We removed this sentence.

Comment: Technical corrections: Page 23041, line 9: change “at large” to “largely” Page 23043, line 3: change “contrains” to “constraints” Page 23047, line 6: change “Similarily” to “Similarly” Page 23048, line 1: “polystyrene” is one word Page 23049, line 3: change “in which” to “to what” Page 23049, line 3: change “asterix” to “asterisk” Figure 4 caption: change "in within" to "within which" Figures 6 – 10: It might be useful to mark the measurement location on these maps. Figures 7 and 8: These appear to be mixed up. Please swap them to match the text.

Reply: Thank you, these were all corrected.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 23039, 2012.