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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The effect of local sources on particle size and chemical composition and their role in aerosol-cloud interactions" *by* H. Portin et al.

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We would like to thank Referee #2 for the comments about our manuscript. All comments have been taken into account. Below are the replies to the specific comments:

Specific comments

Abstract: Parts of the abstract seem to be a bit unclear. In lines 6-9 it is stated "The polluted air masses contained more particles than the clean air masses in all size classes, excluding the accumulation mode. This was caused by cloud processing, which was also observed for the polluted air but to a lesser extent." It is not clear what

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the authors mean by this. A clarification is needed. The statement also does not agree with the results I Table 2. From Table 2 it is quite clear that there also are differences in the accumulation mode between clean and polluted air.

-This part of the abstract was indeed poorly written. We added a few sentences also with some numbers to clarify things. Also, the text now corresponds to the results presented in Table 2. The new text is as follows: "The polluted air masses contained more particles than the clean air masses in all size classes, with average total number concentrations of 2930 and 2000 cm-3, respectively. In general, particle concentrations were lower during cloud events, 1680 and 972 cm-3 for polluted and clean air masses, respectively. An exception to this was the accumulation mode concentration in clean air masses which increased from 146 cm-3 in clear conditions to 349 cm-3 during cloud events. This was caused by cloud processing, which was also observed for the polluted air but to a lesser extent."

Further in the abstract, on lines 14-16 it's stated that for the case study "Clear differences in the total and accumulation mode particle concentrations, particle hygroscopicity and chemical composition during the cloud event were observed." The part about differences in the accumulation mode particle concentrations doesn't agree with the general statement on lines 6-9 that the accumulation mode particle concentrations were similar in both polluted and clean air masses.

-The first half of the abstract summarizes the average results from the analysis of the whole 2010-2011 data set, which indeed did not reveal clear differences in the various parameters between the two air mass classes. The second half deals with the single case study cloud event, which showed much clearer variations. This was not brought out very clearly in the abstract and thus, some modifications to the text were made to highlight this issue.

In the abstract on lines 20-21, it is stated that: "The variable conditions during the event had a clear impact on cloud droplet formation". This also contradicts the statement on

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lines 10-12: The average size and number concentrations of activating particles were quite similar for both air masses, producing average droplet populations with only minor distinctions."

-The same applies here as in the previous comment.

It seems that the general conclusions in the abstract (lines 6-12) doesn't agree with the conclusions from the case study. Please clarify!

-One of the main ideas of the article is that the analysis of the whole data set from the two campaigns can give some idea about what is going on in the different air masses, whereas case studies are necessary to retrieve more detailed information. The abstract was modified to clear things up.

Section 1. Introduction: Maybe the time periods of the two intensive measurement campaigns 20 September-22 October 2010 and 26 September-31 October 2011) could be mentioned already in the last paragraph of the introduction. That would help the reader to follow the presentation in section 2, and understand what you mean with long-term in-situ observations.

-Time periods were added as suggested.

Section 2.2: It seems reasonable to classify cloud events according to visibility, but how is the visibility measured?

-We added a new subsection 2.3.1 in section 2.3 which gives short descriptions about the weather parameter observations at Puijo.

Section 3.2.1: The removal of water from droplets and interstitial particles both from the total inlet and the interstitial inlet could be described a bit more in detail. Is heating to 40 deg C really sufficient to evaporate the water? Are the measurement instruments also at 40 deg C. If not, water might condense on the particles again when the temperature decreases to room temperature. Is the air from the interstitial inlet also heated to 40 deg C? Can you describe the drying process in a bit more detail, in order to make sure

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that all droplets and interstitial particles really are dry before entering the instruments.

-According to our estimations, most of the water should evaporate even in room temperature. For example, evaporation calculations show that in 20 °C and relative humidity of 50% a 15-μm droplet will evaporate completely in 350 milliseconds (e.g. Hinds: Aerosol Technology). The residence time of the aerosol sample in the both sampling lines is more than 10 seconds, the maximum droplet size at Puijo seldomly exceeds 25 μm and the RH in the sampling lines before the instruments is less than 50%. For the DMPS measurements, additional drying takes place when the sample flow is merged with the dried sheath air flow (RH $\sim$10%) inside the DMA tube. Thus, we can safely assume that the aerosol measured by the DMPS is dry and the aerosol sample entering the other devices very likely as well. Some of this information was added to section 2.3.2 (formerly 2.3.1).

Section 3.2.2: If my understanding is correct, the first paragraph, discussing activated fraction, etc., deals with results from the Twin-DMPS. The second paragraph, discussing droplet concentrations, deals with results from the CDP. Maybe the instruments or type of results could be mentioned more clearly. E.g. on page 32144, lines 14-15, you write ". . . within instrumental uncertainty of 20-30%". Is this the uncertainty for the CDP or the Twin-DMPS, or both?

-Yes, the first paragraph deals with the twin-DMPS and the second with the CDP. The instruments are now mentioned in the beginning of the both paragraphs to make things more clear. We estimated the accuracy of the DMPS to be around 10% and added this information also to chapter 2.3.3 (formerly 2.3.2): "For the size range of 20-200 nm, where majority of the cloud droplet formation takes place, the accuracy of the DMPS is estimated to be 10 %, as discussed in Wiedensohler et al. (2012)." Section 3.2.2 was updated: "The average droplet concentrations provided by the CDP (Nd) were 293 and 266 cm-3 for the polluted and clean sectors, respectively. These numbers are comparable to Nact within the instrumental uncertainties of 10 and 30% of the DMPS and CDP, respectively."

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Section 3.2.3 Particle chemical composition: I understand that it might be difficult to draw any conclusions from the results presented, but do you have any ideas? Maybe you can speculate a bit? If not I think it might be good to write that you don't fully understand these results, or something similar. I suppose the mass concentrations should be more or less constant, since there are no major removal processes (as long as it's not raining). The reasons for the observed variations might be mainly sampling and/or instrumental reasons. Or what do you think?

-We added two new paragraphs to section 3.2.3 about the differences between the two sectors and also some discussion about the related uncertainties of this analysis:

"The most significant differences in the in-cloud aerosol composition between the two sectors were the higher concentration of SO4 for the polluted sector compared to the clean sector (1.08 vs. 0.69 μg m-3) and the lower concentration of NO3 (0.19 vs. 0.24 μg m-3). The elevated SO4 may be linked to the local pollutant sources, which produce either SO4 particles directly or then SO2 which is converted into particulate SO4. The more acidic aerosol could also explain the lower NO3 concentration of the polluted sector.

However, based on this analysis, it is impossible to distinguish between the effects of local sources and possible air mass transport from elsewhere on the polluted sector aerosol. Furthermore, as was the case with the particle activation and cloud droplet data discussed in the previous section, also for the particle chemical composition the standard deviations are large, indicating highly varying aerosol properties."

Technical corrections

Page 32138, line 4: Is the inlet PM 1 or PM 10? It says PM 1 but DPM 10 sounds like a PM 10 inlet. Please check!

-Basically it is a PM 10 inlet but it has a nozzle plate for lower cut-off size. This was updated to the text.

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