

Interactive comment on “Hygroscopicity and chemical composition of Antarctic sub-micrometre aerosol particles and observations of new particle formation” by E. Asmi et al.

E. Asmi et al.

eija.asmi@fmi.fi

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First of all, we thank the Referee for her/his useful comments and suggestions. Below, we have addressed these comments in detail.

Comment: The authors should avoid repetitions of possible role of organic compounds. It is very demanding to carry out analysis of organic compounds when aerosol loading is so low, there is no doubt about it. But nearly all information about aerosol organic fraction is derived from bulk PM₁ OC/EC samples with coarse, several days, time resolution. Then this should be clearly stated at one place in discussion or results and avoid repeating paragraphs on possible role of organics, which is mainly speculation.

The fact that organics is an important part of aerosol mass is well established. Discussion being a good example, where in general all possible roles of aerosol organics in aerosol-cloud-climate are listed, but it is all only far reaching speculation for this case study.

Reply: We truly understand this concern and it is indeed demanding to analyse such minuscule masses. In revised manuscript, we have avoided unnecessary speculations on the role of organics and clearly addressed the measurement and analytical uncertainties and how these were taken into account. The uncertainty of the IC analysis is of the order of 10-15 % for all analysed ions and the quantification limit for major ions was 2.5 ng ml⁻¹. The uncertainty of OC and EC results was estimated to be 20 %. The estimated error of the WSOC results is 15 % for atmospheric concentrations < 2 µg m⁻³ and 10 % for concentrations > 2 µg m⁻³. The measured concentrations were above the analytical detection limits and well above (many times) the blank concentrations, which were also subtracted from the presented values. The results regarding both ions and organic carbon were in good agreement with previous measurements made at the site, which further assures the conclusions made. Even though, the exact contribution of organics to the total mass should be interpreted, as you stated, more as a qualitative estimate.

C: Whole discussion of CCN properties carries on again repetition on possible role of organics, but there are no measurements of CCN activity as well as aerosol organic matter speciation. Why is this included at all? The fact that composition is dominated by S-species makes clear point that these particles will be likely good CCN.

R: With this discussion we aimed to address the climatic implications of the results. The measured particle hygroscopic properties together with the supporting chemical analysis can be linked to particle CCN behaviour via the kappa-Köhler theory. We see this discussion more as an attempt to broaden the applicability of the obtained results, although additional data on particle CCN properties should affirm the conclusions.

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C: As far as I understood, authors did not measure aerosol concentration after first DMA, but only after second DMA (page 27308, lines: 5 – 11). I do agree that this should not be a big issue for H-TDMA measurements; however, I do disagree in case of V-TDMA measurements. Authors stated: “. . .the absolute concentration is insignificant. . .” (page 27308, lines: 10-11) and later on that aerosol at Aboa was highly volatile (Chapter 3.5). In case of volatility it is not only about modal peak after heating. If one does not know aerosol concentration after DMA 1, results might show single mode and conclusion about well internally mixed aerosol is easy to derive consequently. But there can be also significant fraction of particles, which volatilizes completely and aerosol is than externally mixed with two populations of particles. How this possibility was considered and treated?

R: You are absolutely right on your comment. We rephrased the paragraph to address for the reader the possibility of underestimating the volatility in case of externally mixed aerosol. However, we believe that the assumption of internally mixed aerosol here was reasonably justified considering the H-TDMA data which showed unimodal growth throughout the measurement period, although it doesn't proof the aerosol was really an internal mixture. If the aerosol was externally mixed, it still doesn't change the conclusion that the aerosol was very volatile, which was the only conclusion we made from V-TDMA measurements.

C: Instead of largely repeating speculations on role of the organic material and at this point questionable volatility data, I would like to suggest addition of DMPS data and discussion of the observed aerosol size distribution, not only nucleation events. It will be useful addition, especially with respect to sizes elected for H-TDMA/V-TDMA measurements.

R: Our main focus in this manuscript was to characterize the particle hygroscopic and chemical properties and detailed analysis of the size distributions was left for following studies. You are indeed right that information on particle size distribution can help in interpreting H-TDMA and chemical data, and gives some proportion to it. There-

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fore, we grouped the measured size distributions based on their modal features into three “typical” clusters. We analysed the prevailing air masses and particle hygroscopic properties corresponding to each of these clusters. The most common size distribution type was characterized by strong Aitken mode, and weak nucleation and accumulation modes. The second common type was a distribution with a strong accumulation mode, typically observed during marine air masses. The third type was the distribution during particle formation, when the particle hygroscopicity was observed to decrease. These three distribution types were plotted and the figure and the following table of the aerosol and air mass properties were added on the revised manuscript. We hope this will give an idea on the aerosol size distribution characteristics during our measurements and its linkages to measured HGF and air mass properties.

C: p 27305, line 2: too many “to”

R: corrected

C: p 27305, line 20: suggestion for change to study natural aerosol processes

R: corrected

C: p 27306, line 9: suggestion for change to . . .the first aerosol hygroscopicity. . .

R: corrected

C: p 27306, line 21: are there any additional checks for contamination besides the SW winds sector?

R: Pollution from the station was clearly visible in the DMPS data, in the H-TDMA data and was also monitored with PSAP. Pollution episodes were only detected during SW winds and data was additionally visually checked to be clean from contamination after removing the pollutant sector data. In chemical samples EC concentration was practically zero at all times insuring that the data was clean and the method of sector control worked well.

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C: p 27307, line 13: How often did you check sheath flows and how stable there were?

R: They were checked every second day together with calibration checks. In the first DMA the maximum deviation from the set value was 0.2 LPM while in the second it was 0.1 LPM. These uncertainties were added on the text.

C: p 27307, line 19: suggestion for change to . . . Particle numbers were measured with a TSI

R: corrected

C: p 27308, line 13: What is the altitude of Aboa station? How might lower pressure during sampling change impactor cut off for various stages?

R: The altitude was 496 m a.s.l. and the atmospheric pressure was between 920-940 hPa. This small deviation from standard pressure should not change the cut off sizes significantly.

C: P 27309, line 11-23: did you check the real cut off of the unit used for OC/EC sampling with DMPS for example?

R: Berner has calibrated the inlet cut off with nominal flow rate and we calculated the change of cut off for the flow rate used. There was no reason to believe that the calculation would not apply at the given conditions, and therefore, we didn't assure this with an additional measurement.

C: 2.2.2: Chemical sampling and analysis: Authors should discuss here all possible errors and uncertainties, precision and accuracy, so the reader can assess results discussed later on properly.

R: We added discussion on the uncertainties, and on the assurance of the quality of the data. This issue was mainly covered in our answer to the first question.

C: P 27310, line 9-10: how do you define "wet diameter". Would not be more appropriate use the term "ambient"? It is often dry in Antarctica, so "wet diameter" might be

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misleading

R: You are right, we corrected the “wet” to “ambient”.

C: P 27312, line 6: these data are “weather conditions” not “climatic conditions”. R: corrected

C: P27312, line 25: does it mean that aerosol volume is PM1 volume? If so, it should be clearly stated.

R: Yes, this was added.

C: P 27313, lines12 -21: how is recent marine influence defined? Did you consider also altitude in you trajectory analysis?

R: Recent marine was determined based on general characteristics in Fig. 3, and it corresponds to air masses which had come from the ocean within about 24-h. The trajectory analysis in Antarctica is very uncertain and individual trajectories can not be considered as too reliable. Therefore, air masses were classified as recent marine only if the same feature had remained for over half a day and trajectories calculated for different heights showed a similar behaviour. Because of the short measurement period the trajectory height was not considered as the statistics would have been poor. However, we are planning on analysing the characteristics of size distributions over several seasons where the trajectories can be considered in more detail.

C: P 27314, line1: can you give numbers by how much higher? Is it statistically significant?

R: The statement “higher” refers to the visible trend seen in figures 3 and 4. We would prefer not to give exact numbers because the number of chemical samples (12) is too low to make statistical analysis of the correlation and also, because the individual trajectories contain large errors especially in Antarctica making statistical analysis based on the low number of trajectories slightly questionable. However, because of your request, we calculated the correlation and found a decreasing trend in MSA/nss-sulphate

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as a function of the trajectory propagation time from the open ocean. Decreasing trend was found regardless of the trajectory height used and/or the method of averaging the trajectories to match the times of the chemical sampling. Most of these correlations were non-significant ($p > 0.05$) but depending on the method of averaging the trajectories also significant correlations could be obtained.

C: P27314, line 26: How do you define daytime? Some thresholds in solar insolation?

R: Daytime here refers to the time of highest solar radiation. Despite the constant sunshine, the daily cycle in solar radiation is definite as seen in Fig. 2.

C: P 27315, lines 1-16: Do these correlation have any meaning and statistical significance with respect to very low number of samples?

R: Majority of them show p-values much less than 0.05. We added a mark on the table on those values which do not meet this criterion. For example, the HGF of 10 nm was observed only occasionally and therefore, statistically significant correlations could not be determined with most of the parameters.

C: P 27319-20: why there is no analysis of nucleation event on 20 Jan, which is different from next day “banana” nucleation

R: On Jan 20 only the growing mode was detected while the actual nucleation had occurred earlier on that day. As nucleation was not detected, also the nucleation rates could not be determined.

C: P 27320, line 19: particle dynamics and/or different air mass origin

R: True, corrected.

C: p 27322, line 14: these elements are also emitted from the ocean in a form of sea spray aerosol

R: This was added

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