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Atmospheric
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Discussions

Interactive comment on “LACIS-measurements and parameterization of sea-salt particle hygroscopic growth and activation” *by* D. Niedermeier et al.

D. Niedermeier et al.

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We thank referee 2 for his/her comments, which we respond to below:

General Comments:

1a. "On page 4, 4th paragraph, 2nd column: repeat the sentence ’Sea salt particles exhibit reduced growth compared to NaCl ...’ in the abstract. This is an interesting finding."

The following sentence concerning the reduced hygroscopic growth of the sea-salt particles compared to NaCl particles is inserted into the abstract: ’The measurements showed that the sea-salt particles exhibit a slightly reduced hygroscopic growth com-

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Discussion Paper

pared to NaCl particles.'

1b. "Given the observed chemical composition of the sea water samples, is the reduced growth consistent with the differences in chemical composition? Or does there need to be some unobserved insoluble component invoked to explain the reduced growth?"

Motivated by the referee, we performed some additional model calculations using the Pitzer model (Pitzer and Mayorga, 1973) for simulating the hygroscopic growth of the sea-salt particles. In these calculations only the inorganic components, found in the samples, were considered. These model calculations yielded a hygroscopic growth for the sea-salt particles that was slightly larger than the measured growth. However, this discrepancy only amounts to maximal 5% in the wet diameters at the highest measured RH. This, compared to theory, slightly reduced growth may be caused by small amounts of e.g. less hygroscopic organic material and uncertainties in the derived density. Of course also insoluble components, as suggested by the reviewer, might by a reason. The following sentence was added to the manuscript: 'This small difference between the measurements and calculations may be caused by small amounts of additional less hygroscopic organic or insoluble substances in the sea-salt particles and uncertainties in the calculated density of the sea-salt particles.' In addition to this sentence, the text was changed in sections 4 and 5 to describe the model and its results.

Specific Comments:

1. "On page 2, last paragraph: were blanks also analyzed to verify no contaminants were introduced (for example ultra pure water in same containers used to store the sea water, filled at same time as sea water samples taken, and analyzed simultaneously)."

No blanks were analyzed. However, if there were significant amounts of slightly soluble or insoluble substances in the containers and therefore (maybe) in the generated particles, the hygroscopic growth of the sea-salt particles would not have been as strong as the measurements suggest. The small influence of contaminations becomes obvious from the modeling efforts described above.

2. "On page 3, when atomizing and drying the sea water-derived particles, if supersaturated sea salt solutions are created during the drying process are the pairings between the various elements changed due to solubility issues?"

Since the different elements in the created particles dissociate in the saturator / in the flow tube, the pairings of the various elements is of minor interest for the hygroscopic growth and activation. This holds especially true as in the applied model (see above) only the measured ions are considered.

3. "On page 3 - how 'dry' is 'dry'? How did you verify that the particles being selected by the DMA were in fact free of water? Were there any droplet shatter patterns on the ELPI grids that may have indicated some water remaining on the particles?"

The RH after the diffusion dryer is below 10%. Additionally, the ELPI measurements showed no droplet shatter patterns, so there was no evidence that water remained on the particles. Again, the results of the applied model suggest only a small or negligible influence of water remaining on the dry particle.

4. "On page 4, 2nd paragraph: If the sea water contained organic species that potentially formed monolayer coatings on the droplets, do you think a 2 second residence time in LACIS would bring them to their equilibrium size?"

The measurements showed that the sea-salt particles exhibit a large hygroscopic growth and this is just slightly reduced compared to the theoretically predicted growth. That means that the droplets are most likely not completely covered by an organic monolayer. In the manuscript we inserted a sentence that the slightly reduced hygroscopic growth compared to the theoretically predicted growth is maybe caused by small amounts of less hygroscopic organic or insoluble substances.

5. "On page 4, 2nd paragraph, 2nd column: would larger sea salt particles also be of interest? 185 nm still seems slightly small compared to 'typical' ambient sea salt particle diameters."

One of the main focuses of this study was to explore the possibilities to achieve of CCN-closure. Therefore, particles in the same size range were investigated concerning their hygroscopic growth and activation. The result of the activation measurements was that sea-salt particles with a Dme around 100 nm require a supersaturation of about 0.1% for activation to become cloud droplets. Such supersaturations are a) frequently reached in the atmosphere and b) very much at the lower limit of supersaturation controllable in CCN counters and LACIS. Therefore investigating particles larger than 185 nm is according to our opinion a) of less importance in the context of this manuscript and b) beyond (our) existing experimental capabilities.

6. "Do you think ammonium sulfate might also require a shape factor correction? If you repeat the shape factor tests conducted on the sea water and sea salt samples with ammonium sulfate, do you observe a shape factor of unity?"

Following the referee's suggestion, ELPI measurements were repeated for ammonium sulfate particles and a shape factor of about 1.04 was found (as stated in literature (Zelenyuk et al., 2006)). Therefore the whole data analysis was redone. The result was that changes in RH due to the shape factor correction are small. For high RHs, the uncertainty in LACIS amounts to +/- 0.3% in RH absolute. The influence of a shape factor of 1.04 vanishes within these uncertainties. Changes due to the shape factor correction of ammonium sulfate particles were performed in Sections 5, 6 and 7 and in Appendices A and B.

7. "On page 2, 2nd paragraph, the sentence starting 'In this context...' is difficult to follow and should be rewritten. The following sentence should read: 'To explore the affect sea water chemical composition may have,...' "

The sentence on page 2, starting with 'In this context...' was rewritten: 'In this context it is also relevant to know, if possible differences in the chemical composition of the oceans influence the hygroscopic growth and activation behavior of the generated sea-salt particles. To explore the affect sea water chemical composition may have, sea-

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salt particles from three different sea-water samples were investigated regarding their hygroscopic growth and activation to cloud droplets.'

References

Pitzer, K. S., and Mayorga, G.: Thermodynamics of electrolytes. II. Activity and osmotic coefficients for strong electrolytes with one or both ions univalent, J. Phy. Chem., 77, No. 19, 2300–2308, 1973.

Zelenyuk, A., Cai, Y., and Imre, D.: From agglomerates of spheres to irregularly shaped particles: Determination of dynamic shape factors from measurements of mobility and vaccum aerodynamic diameters, Aerosol Sci. Technol., 40, 197–217, 2006.

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