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Atmospheric
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Discussions

Interactive comment on "Hygroscopic growth of sub-micrometer and one-micrometer aerosol particles measured during ACE-Asia" *by* A. Massling et al.

A. Massling et al.

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Title: Hygroscopic growth of sub-micrometer and one-micrometer aerosol particles measured during ACE-Asia

Comment on the general comments of referee 1:

Measurements dealing with physico-chemical properties of aerosol particles of the Asian plume are very scarce. The authors will try to find additional references to compare our results with other studies. We only think of studies like Aerosols '99 and INDOEX, which show other comparable data sets downwind of major continental, anthropogenic source regions.

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Unfortunately, a detailed chemical composition is always the problem when interpreting hygroscopic growth data. In some cases, a size-segregated chemical composition is available, but the data do not match the time resolution of the hygroscopicity data. Also, any kind of impactor data and filter data only provide bulk analysis resulting in the missing information of the state of mixture. Chemical information specific to only the Aitken mode are not available for this study.

The reviewer is right, that we comment on the chemical composition, perhaps too strongly, based on only hygroscopic growth data. (Note that we do preface most of statements as assumptions. This is not speculation!) The authors will change some of the specific explanations following the comments on the specific comments listed below in a revised version of the manuscript.

Our main point is the presentation of the size dependent growth factor data, relative number fractions in the growth modes and the extension into the supermicrometer size range. And this is done in the context of particle size, air mass, and location. What we can say about the chemical composition of the particles from the growth factor is that, in most of the measured cases, the fraction of elemental carbon, insoluble organic carbon compounds or slightly soluble compounds is small compared to inorganic compounds such as ammonium sulfate or (partially neutralized sulfates) and nitrates. The organic compounds that are there must be highly oxidized, likely acids or diacids.

Comments on the specific comments of referee 1:

Abstract line 24: Unfortunately, we do not have a detailed chemical information in this specific size range. It is correct, that ammonium sulfate is only one possible explanation of the observed growth factors. Please also have in mind the uncertainty of the measurement. A revised version of the abstract will consider the mentioned possibility of non-neutralized sulfate. Sodium chloride particles mixed with organic compounds due to sea spray processes should have growth factors around 2.0 and higher. Please note, that the comment in the abstract deals with particles of a size range between Dp

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= 50 and 350 nm. Here the growth is also dependent on the Kelvin effect.

Page 12271, line 23: You are right, this will be added in a revised version of the manuscript.

Page 12273, line 2-3: Thank you, I agree, this makes it more clear. This will be changed in a revised version of the manuscript.

Page 12273, lines 7-8: Actually, there are a couple of RH measurements in the system. But, for the regulation control of the aerosol sample flow and the sheath air flow, we can only use one sensor that we trust in. So, we use the NaCl calibration, which is - in our view - the best method to get the real RH value during the measurements.

Page 12273, line 15-19: You are right, I also see no way to correct for multiple charges in TDMA measurements. The expression inversion is maybe not the best way to describe what is done here. In a revised version of the manuscript, we will add a few sentences to avoid that the reader thinks we would correct for multiple charges which we do not do.

Page 12273, line 29: We know that this is always a problem, especially if the data is noisy. The fitting then can sometimes be a highly subjective job. If clear modes are not really observed, we always propose other parameterizations than mode fitting. In this study, we averaged distributions as we described in the manuscript. These distributions resulted in those showing clear groups of different hygroscopic behavior. One figure is added to show examples of some hygroscopic growth distributions.

Page 12274, line 2-3: Good comment. It is number mean diameters. This will be mentioned in a revised version of the manuscript.

Page 12274, Eq. 2: Right, our indices were confusing. This will be changed in a revised version of the manuscript.

Page 12274, line 11: The measurements were done for particles with a dry size of 150nm. Dry sodium chloride particles are understood to be cubic. All issues mentioned

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in your comment were considered when calculating the DMA RH from the experimental growth of the sodium chloride particles. In future studies, we go over to use ammonium sulfate particles. Their growth is smaller, but they are understood to be spherical at dry size.

Page 12274, line 14-15: The deviation from the nominal RH was random for all events. This may be due to temperature fluctuations within the container depending possibly on the number of persons in the container. For systematic deviations, there are some possibilities to correct the growth factors as you mentioned. But, as discussed before, based on our quality criteria, we have sorted the data and averaged growth distributions sharing the same parameters during specific time periods. For this reason, as we had deviations in both directions (higher RH and lower RH), fluctuations will also cancel out.

Page 12274, line 17-18: With ammonium nitrate and sodium nitrate you have picked out extreme cases in growth differences at 90% RH. But I agree, that these species have atmospheric relevance possibly also in our study. We will change our estimate in uncertainty of hygroscopic growth factor based on your comment in a revised version of the manuscript.

Page 12276, line 4: Right, this is unclear in the manuscript and will be left out in a revised version of the mnauscript. The comparison for this campaign was based on the aerodynamic particle size distributions measured with the second APS only, at 30 and 90 % RH. In normal operation, comparison of the particle diameters measured in dry and hydrated states with the two APS units yields the aerodynamic growth factor gfaero. During ACE-Asia, particles with an initial dry size of Dp=1 μm were selected in the DMA, and subsequently sized at 30% and at 90% RH using the second APS unit. The measurement RH was changed periodically, with individual measurements at a specific RH lasting for 15 min. The first APS unit, measuring the size distribution of the particles selected by the HAF-DMA in their dry state, showed sizing problems during this campaign. Therefore, the growth factor was calculated by using the measurements from the second APS unit alone, by comparing the aerodynamic particle size measured at 90% RH to the one measured at 30% RH. We may also omit the dry particle number size distribution in Figure 7b showing only 30 and 90% distributions, as they are the ones mentioned in the paper.

Page 12276, Eq. 3: This is right. This parameter can vary based on density fluctuations. It was derived by measurement of the dry (30%RH) scans taken regularly during the cruise.

Page 12276, lines 13-16: No, the H-DMA-APS does not need any inversion algorithm. The APS provides an aerodynamic particle number size distribution. Possible doubly charged particles leaving the High Volume Flow DMA could clearly be identified in the further data analysis.

Page 12277, line 6-7: Setting the growth factor of nearly hydrophobic supermicrometer particles to 1 is somewhat arbitrary. This value has to be treated as a qualitative one, stating that there is practically no growth. Because of uncertainties, calculations of the Stokes equivalent growth factors for nearly hydrophobic particles were sometimes undershooting the value of 1, which is not realistic. In contrast to this, the number fraction of nearly hydrophobic supermicrometer particles is a quantitative value and can be calculated as described in the manuscript. This will be stated more detailed once more in a revised version of the manuscript.

Page 12277, lines 25ff.: This is right, if the sulfate is not fully neutralized in the marine case, this could be a good explanation for the small overshooting of measured hygroscopic growth factors compared to those of ammonium sulfate. A revised version of the manuscript will take this possibility into account.

Page 12278, lines 11-13: This is good additional information and will be added in a revised version of the manuscript.

Page 12278, lines 18-24: We do not agree in this point. Some of your suggestions

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are unlikely at best. First, we do not see atmospheric processes producing mixtures of soot and sodium chloride or dust and sodium chloride particles. Sodium chloride particles did not occur in measurable number fractions at Dp = 350 nm in this air mass and therefore it is most unlikely that these particles occur at smaller sizes since they are produced by sea spray processes. Second, dust particles are excluded based on the air mass analysis and size range in this case. The authors do not exclude any other possibilities you mentioned. Here, soluble substances could be sulfate or nitrate salts mixed with less or non hygroscopic material like soot or organics.

Page 12279, lines 2-4: This is good additional information and will be added in a revised version of the manuscript.

Page 12279, lines 10-11: This is a good explanation, which is independent of aging times. This second explanation will be added to the first explanation in a revised version of the manuscript.

Page 12279, lines 21-23: Again, we would not expect these particles to be a mixture of sodium chloride and organics. Supermicrometer measurements do not show pure sea salt particles. Thus, we do not expect to find aged sea salt particles in the submicrometer size range.

Page 12279, line 23: We agree, this is better and will be changed in a revised version of the manuscript.

Page 12279, line 23-30: In our set up, the particles were instantly dried to values smaller than 5 % RH. But I agree, that particles could partially been neutralized to ammonium bisulfate. This possibility is not excluded in our considerations, but will be emphasized in a revised version of the manuscript.

Page 12280, lines 4-9: This is a good point. One possibility could be, that supermicrometer particles had a different origin than submicrometer particles. Looking at the different growth factors, it looks that supermicrometer particles were fully neutralized.

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A second possibility could be the higher probability for supermicrometer particles for cloud processing leading to fully neutralized particles.

Page 12280, lines 27-29: This is right. Sea salt as dust particles have their maximum in number between 1 and 10 micrometers. The fractioning of these two groups depends on the individual air mass, on the emission ratio of the specific particles, and their probability to be washed out or sedimented during transport. An explanation for the specific number fractioning in this case is not possible.

Page 12281, lines 7-10: Please see comment on Page 12278, lines 18-24. Also, we do not imagine, that for this period aged dust particles could explain the mentioned growth factors. Aged dust particles of dry sizes of 250nm growing up to a factor of 1.5 must originate from 100nm dust particles. From our knowledge, dust particles have not been observed in this size range. This leads us to the assumption, that these particles were of anthropogenic origin.

Page 12281, lines 10-14: We think, they can. Soot particles are known to have their emission maximum in number between 50 and 100nm in dry diameter. If these particles were soot particles, we would expect them to appear for smaller sizes, too. Here, this is not the case. Nearly hydrophobic particles did not appear for dry sizes of 50 and 150nm. Furthermore, a nearly-hygrophobic class was not observed in air masses from pollution source regions alone.

Personal interest: This is an interesting question. I do not see the chance to get a correlation between the occurrence of sea salt particles and their maximum in number and the described parameters determined here because of a lack of data. We know, that there exist models describing the sea spray process and the production of particles in different size ranges. Basically, particles in three different size ranges can be produced via sea spray processes. In my view, a monitoring station close to a coast line providing hygroscopic growth data could possibly provide information about a link between wind speed or other meteorological data and the occurrence of sea salt particles in different size ranges.

Comments on Tables and Figures:

Tables A1 - A5: Thank you for the comment.

Figures 3-6, 8: This is the accuracy of the reported values. This will be added in the figure caption in a revised version of the manuscript.

Figure 7 caption: This is a good comment. The Figure caption will be extended by your comment in a revised version of the manuscript.

Figure 7b: This is not possible since there is no clear dependence between the Stokes diameter and the aerodynamic diameter. This is because the density of a particle is changing with increasing amount of water on the particle depending on the particles hygroscopicity.

Comment on technical corrections:

Thank you for your technical corrections. All technical corrections will be considered in a revised version of the manuscript.

Acknowledgement

In general, I want to thank the referee for his interesting questions and several possible hints to increase the quality of the manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 12267, 2006.