Atmospheric Chemistry and Physics Discussions

Interactive Comment

Interactive comment on **"Latitudinal aerosol size distribution variation in the Eastern Atlantic Ocean measured aboard the FS-Polarstern"** *by* **P. I. Williams et al.**

P. I. Williams et al.

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Response to referee 2

"The authors do not mention whether they had any control device to eliminate pollution from the ship itself, or whether the periods affected by ship-derived pollution were identified somehow in their data/error analysis. This is important because ship emissions are known to contaminated particle number size distribution data in marine environments."

One of the parameters recorded by the ship's data logging system was the relative wind direction and this was used to filter the data. The sample inlet was upwind of the exhaust, and so a criteria was used where by any data associated with a relative wind

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direction from 135 to 225 degrees was rejected. This detail has been included in the revised manuscript:

"One of the parameters recorded by the ship's data logging systems was the relative wind direction. This allowed the data to be filtered for potential contamination from the ship's exhaust. Data that was associated with a relative wind direction from 135 to 225 degrees was removed, as it could contain aerosol from the main exhaust."

"The authors spend some time in discussion the modal structure of the measured size distributions. Here I would like to see some more comparison with other similar measurements. In addition to the work by O'Dowd et al. (1997), marine aerosol number size distributions have been discussed comprehensively in the studies by Covert et al. (1996, JGR 101, 6919-6930), Heintzenberg et al. (2000, Tellus 52B, 1104-1122) and (2004, Tellus 56B, 357-367). Furthermore, in the work by Koponen et al. (2002, JGR 107(D24), doi:10.1029/2002JD002533) the modal structure of the submicron aerosol was measured at approximately the same route and same time of the year as in this manuscript"

The Koponen et al. paper does provide a very useful comparison to the modal structure of submicron aerosol, not only because it covers an almost identical route, but also because it uses nearly identical methodologies for measuring the aerosol size distribution. There are some factors that need to be highlighted that introduce an uncertainty in making a direct comparison. However, the Polarstern data have been reanalysed to fit log-normal distributions to the data enabling geometric mean and a standard deviation to be presented. The geometric mean diameter of the accumulation and Aitken modes has been updated in fig 5 which was previously based on fitting a quadratic to the data and looking at the second derivative to determine the peak diameter. One of the major differences between this data and the Koponen et al. is the relative humidity in the DMAs. The relative humidity was kept at less than 20% in Koponen et al., where as in this data set it was keep at ambient. Comparing data from similar areas (denoted Marine Air A in Koponen et al), the Aitken geometric mean diameter is given as ~40nm

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in Koponen et al., whereas 64nm was more representative of the current dataset. For the accumulation mode, Koponen et al. reports a value of ˜176nm whilst that in the current study is 220nm. At 70% (80%) RH, a 40nm pure sea salt particle will have a growth factor (GF) of 1.78 (1.91) and for 176nm pure sea salt, a GF of 1.81 (1.99) (ADDEM model, David Topping, private communication). Using these growth factors to grow the Koponen et al. Aitken mean mode diameter yields a mean modal diameter of 71.2 - 76.4nm, which is similar to the sizes in this study. It is possible that the particles were not entirely efflorescenced in the drying process reported in the Koponen et al. study. A better comparison will be to compare Dp(Williams) with Dp(Koponen) x GF70-80%/GF20%. The ADDEM model predicts a GF20% of 1.38 and 1.39 for a 40nm and 176nm particle respectively. This yields an Aitken mean diameter of 51.6 - 55.4. It is likely that the particles in this size range are not pure sea salt and will have a different GF and the value of GF20% is likely an over estimation as the model is at the limit of its capability at that RH. These factors complicate the comparison. For the accumulation mode, performing a similar task and multiplying the Koponen et al. size by the GF yields an 'ambient' aerosol size of ˜ 318 - 350nm, which suggests that the geometric mean diameter is substantially larger than that reported by this study. Comparing Dp(Williams) with Dp(Koponen) x GF70-80%/GF20 as above, yields an ambient wet size in the range of ˜229 - 252nm, which is closer to the measured ambient conditions experienced on board the Polarstern. It is important to remember that at RH = 20%, the ADDEM model is at the limit and some of the assumtions therein may be invalid and that GF20% is likely to be significantly lower. However, this highlights a potential difference between the two data sets, as a shift of 30-50nm or more, will have important implications on the CCN properties and it will also increase the condensational loss rate calculations. Similar results are reported in Heintzenburg et al. 2000 which compiles data from a variety of campaigns to generate a 15 x 15 degree grid of marine aerosol data. In this review, a criteria is set that only data with an RH $<$40% is used, hence direct comparison with this data set is problematic. Comparing data from 0 to -15 degrees, the Aitken (accumulation) mode diameters are 47nm (170nm) and 66nm

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(222nm) for Heintzenberg and this study respectively. Covert et al. 1996 present data from the mid-Pacific and describe the sub-micron aerosol size distributions recorded using a DMPS system. Although not stated explicitly, one assumes that these measurements were made a RH < 40% as the data is included in the Heintzenberg review article. This makes for an interesting geographical comparison. For latitudes similar to those that this study measured, the Aikten mode is reported as having a mean diameter from 30 - 60nm (figures 3a and b in Covert et al.), and for the accumulation mode from 150 - 250nm which is much closer to the diameters recorded during this study at ambient RH. This would imply that the ambient size (at ambient RH) of the accumulation mode aerosol is higher in the mid-Pacific than in the mid-Atlantic. It is more difficult to perform a comparison of the number concentrations. This is complicated by two factors: Firstly, in the cited articles, the concentration is not reported with an associated wind speed. If one is to compare a mean number concentration, then that needs to be reported along with a mean wind speed. Furthermore, the Heintzenberg review states that there are gaps in the data both for compositional information and number concentration at high wind speed. This re-affirms the comments made in the response to reviewer 1 that more work is needed in this area. Secondly, as stated in the paper, there is an issue with the DMPS data at moderate to high wind speeds. The revised article includes these comparisons and a full discussion as above addressing the potential effects of relative humidity on the measurements.

"Calculation of the trace gas loss rates (section 3.1) is interesting, especially since the authors have practically measured the whole particle size range relevant for this loss. There are a few points that might deserve some additional discussion. First, it is not mentioned which value of the accommodation coefficient has been assumed in Figure 9. This value can have a large influence on the "size distribution" of the loss rate depicted in Figure 9. Second, the authors should mention that the distribution given by Figure 9 might be affected by the fact the either the accommodation coefficient or the heterogeneous reaction rate might depend on particle type (and size), i.e. they could be different for supermicron and less acidic sea-salt particles than for submicron,

sulfur- containing and more acidic particles. Third, the loss rate is interesting not only because of heterogeneous marine chemistry but also because of other processes such as new particle formation. As we know, new particles formation in most marine environments seem to be rare, possible because of moderate sources of aerosol precursors combined with relatively large condensation sink caused by sea-salt particles."

In fig. 9, the uptake coefficient of HOI, 0.03, was used. This will be stated explicitly in the text.

The reviewer is correct in the statement that the actual value of the uptake coefficient will depend on the chemical nature of the aerosol particle. The uptake coefficient is a combination of the reactive uptake of the chemical species, the accommodation coefficient and any inhibition of uptake caused by the surface. These will depend on the chemical nature of the aerosol particle, which will vary across the size range. The values of the uptake coefficient where chosen to illustrate the effects on the different moments within the aerosol size distribution. In the case of uptake coefficient = 1, the loss rate is limited to rate at which species X can diffuse to the surface, and the loss rate has a relatively higher dependence on the smaller sizes. In the limit where the uptake coefficient = 0.03, the loss rate is limited by the uptake of the available surface and therefore has a relatively higher dependence on the larger sizes. In the revise manuscript, this will be included in the discussion as will an additional trace in Fig 9 which shows the lose rate with uptake coefficient = 1 as well as 0.03 for a wind speed of 8 m s-1. The loss rate may have an effect on the potential for nucleation in the MBL. Referring to Fig 8, this shows that there is approximately a factor of 10 variation in the potential lose rate from the 3rd of Nov 2003 onwards. Therefore, if the factors that control this variability, such as wind speed, have less than a factor of 10 influence on the production of potential precursors, then the increased potential loss rate will be likely to further suppress nucleation. If however these factors increase the available precursors at rate higher than the potential lose rate, then this will potentially overcompensate for the increased aerosol surface area. Obviously the effect on particle formation will be

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dependent on the non-linearity of the nucleation process and precursor availability (e.g. gaseous oxidation needs to be considered in the production of nucleation precursors) and a direct relationship between the nucleation potential and the difference between the windspeed enhancement of condensational sink and precursor formation may not be evident. Both the chemical nature of aerosol and potential effect on nucleation will be discussed in section 4 of the revised manuscript as detailed above.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 12865, 2006.