Atmos. Chem. Phys. Discuss., 6, S7023–S7027, 2007
www.atmos-chem-phys-discuss.net/6/S7023/2007/

Atmospheric Chemistry and Physics Discussions

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.

Received and published: 5 March 2007

Firstly,the authors would like to thank all the referees for there time and effort in reviewing this article. I will address all the points raised here and where necessary in the revised manuscript.

Referee 1:

General comments

"I have some concerns with the 'observed' mode at 0.6 um , as detailed below."

I have addressed this in detail below.

"The discussion on the loss rate of trace gaseous compounds (section 3.1) seems

artificial: all the authors do is take the particle size distribution for the evaluation of eq. 3. Figures 8 and 9 show this. Figure 8 shows the variability of the integrated volume size distribution, displaced by the uptake coefficients, and Figure 9 shows in principle a modified (by the term in parentheses in eq. 3) mass size distribution. Hence it is no surprise that the maximum loss rates occur at the peaks of the volume size distribution. There is no comparison with data, and hence the discussion remains not conclusive, as regards the actual loss rates and the uptake coefficients (anything between 0.03 or 1?). I suggest to either shorten this section or delete it completely and include this assessment as part of the conclusion, i.e. the current last par. on p. 12880."

Firstly, the author is incorrect in stating that Figure 8 shows the integrated volume size distribution variability displaced by the uptake coefficients. The calculated mass transfer rate calculated using equation 3 would scale directly to a surface area if there were no diffusional limitation. Since there is a diffusional limitation to mass transfer to larger particles, and this limitation is more pronounced when the accommodation coefficient is high, the actual mass transfer rate scales to somewhere between the first and second moments of the distribution dependent on the size distribution shape and the uptake coefficient. The mass transfer rate is therefore not directly related to the volume distribution. Figure 8 shows how the available condensation sink varies with time. Figure 9 does not describe a modified mass distribution - it describes a mass transfer distribution, showing which parts of the size distribution contributes most to the condensation sink as a function of particle size. The condensational loss rate, kt, has two controlling factors: the aerosol number size distribution and the uptake coefficient. The focus of this section of the paper was to examine the limits and variability of this lose rate caused by the changing aerosol microphysics. It was not to determine actual loss rates. Figure 8 shows the total potential loss rate (i.e. condensational sink) summed over all sizes. This can be referenced with any gaseous species with an uptake coefficient between 0.03 and 1. Figure 9, which represents the lose rate in eq. 3 as a function of size at an uptake coefficient of 0.03, illustrates the major sinks for the uptake of different gases as a function of wind speed. This analysis provides

ACPD
6, S7023–S7027, 2007

EGU

the reader with a reference condensational sink variability for measured marine aerosol size distributions. I believe that shortening or deleting this section would be detrimental to readers interested in reactive uptake of marine gases.

“p. 12868, line 3: suggest to include Clarke, A.D., S.R. Owens and J. Zhou, 2006. An ultrafine sea-salt flux from breaking waves: implications for cloud condensation nuclei in the remote marine atmosphere. J. Geophys. Res., 111, D06202, doi:10.1029/2005JD006565, 14 pp.”

This reference will be included in the revised manuscript.

“Abstract, line 11: what is the range of wind speeds?”

The wind speed ranged from ˜1.7 to 14.7 m s-1. This will be included in the abstract of the revised manuscript

“p. 12873, line 14: the mode at 0.6 um is hard to see in Figure 4, at the higher wind speed there is a hunch. Would it be more convincing if plotted as dV/dlogD (in fact this is doen in Figure 9)? See also next comment:

p. 12874, line 8: I think this mode is not very noticeable. Is this mode really significant. In view of standard deviations and errors? Especially in view of the concern expressed by the authors in the par starting at line 30, as regards sampling errors. Apparently the DMPS system had a sampling problem at higher wind speeds. Could this cause an apparent decrease in the concentrations smaller than 0.6 um, leading to an apparent increase resulting in the 0.6 um mode?”

Addessing these comments together: the size distributions are presented in number rather than volume space to enable straightforward comparison with number distributions in other published work. Note: Figure 9 is not a mass distribution; it is the condensational loss rate as a function of size (evaluation of eq. 3), see response to previous comment. It does show more clearly the appearance and development of the mode at 0.6 um, but cannot be used to compare with other work as it is neither number

ACPD

6, S7023–S7027, 2007

nor mass. The mode at 0.6 um is significant. Figure 4 shows the standard deviations as error bars, indicating that the concentrations are distinct at each chosen wind speed, however the authors acknowledge that due to the log scale and the several orders of magnitude on the graph, it is hard to see. The problem with the DMPS did not affect this mode, as this size range was measured by the GRIMM OPC (which measured to 0.3 um), and not the DMPS. Sizes down to 0.3 um are believed to be correct at higher wind speeds. Where the DMPS was used below this size, the number concentration was affected at the higher wind speeds as highlighted in the paper.

"p. 12875, line 24: the apparent plateau in the wind speed dependence is a concern, especially for small data sets. Usually there are only few data for the highest wind speed. When data are available for wind speeds higher than in the data set presented, it is often observed that there is no plateau. The curves in Figure 7 which have error bars on them seem to be all within experimental error. Hence these data are not conclusive as regards the shape of the wind speed dependence. In particular I object to the conclusion that based on this data, which deviate from other data sets, is concluded (p. 12879, line 10-15) that 'the data suggests that there is a limit to the mechanical generation of film and jet modes in the open ocean'."

The reviewer makes a valid comment that within the errors, a function as defined in eq. 1 can be fitted to the data set within the standard deviation of all points. However, the fit is significantly better using a sigmoidal function, with the residual to the fit being less, as shown below, especially at high wind speeds.

Wind Speed 2.3, 4.2, 6.0, 8.1, 10.0, 12.0, 13.7

Sigmodal -0.35, 0.35, 0.07, -0.13, 0.08, 0.02, -0.04

O'Dowd-fit 0.36, 0.67, -2.08, -0.75, 2.78, 2.19, -2.24

It is acknowledged that this statistical improvement indicates, but does not prove, that there is a mechanical limit to the production of the particles at high wind speeds. More

ACPD

6, S7023–S7027, 2007

work is clearly needed on this subject. The revised manuscript now states:

“a sigmodal function represents the data more accurately, indicating a limit to the number of particles produced at high wind speed, but a function as defined by O’Dowd et al could be fitted within the errors. It is unclear whether there is such a limit, but the suggestion of one requires further investigation.”

“p. 12876, line 2: I am not sure that these particles are likely sea salt. Recent evidence shows that other materials contribute a large fraction to the sea spray aerosol (e.g.,O’Dowd et al., 2004). The term ‘sea spray’ may be better here. In the discussion the authors also point out that there may be additional organic material present in the particles (p. 12878, line 20)”

The term sea salt will be replaced with sea spray.

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