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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #3

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Summary: The paper describes the use of the LACIS apparatus to develop a parameterized equation describing the hygroscopic growth of sea-salt particles.

LACIS uses a DMA to select a monodisperse collection of diffusion dried particles obtained from atomized sea-salt solution. The dried monodisperse aerosol is exposed to varying relative humidity $>$ 80% and the size of the resulting droplets can be measured with an optical particle counter. LACIS requires calibration using sodium chloride, ammonium sulfate, and polystyrene latex sphere standards.

Köhler theory with both the Kelvin and Raoult terms, which describes this process, requires the diameters of the dry particle and the resulting droplet as well as several

parameters that are characteristic of the salt. Since none of the salt-characteristic terms are known, they are combined into an empirical term called the ionic density, rho_ion. The ionic density was varied until the droplet diameter calculated by Köhler theory matched the observed diameter (shown in figures 3 and 4). The results of these calculations are reported in figs 7 and 8.

Next the activation of cloud formation by sea salt particles under super-saturated conditions was measured (figs 5 and 6) in order to determine the critical diameter (Table 3 and Figure 9, filled squares). The ionic density were then used to derive the parameterized Köhler equation given by equation 6, which can be used to predict the critical diameter as a function of supersaturation (open squares, figure 9).

General comments: The parameterization of Köhler droplet growth from sea salt aerosol will be useful to modeling cloud formation in marine environment. The authors have made a series of very careful measurements with high precision instruments. I have confidence in the measurements of the droplet diameters that were produced. However, I have significant concerns about the accuracy of the measurements of the diameters of the dry particles. The mass equivalent diameter of the dry particles was determined by applying a cubic morphology shape factor to the aerodynamic diameter determined by the differential mobility analyzer (DMA). Determination of the shape factor rests heavily on two unsupported assumptions:

1. That the sea salt aerosol emerging from the diffusion drier is in fact completely crystalline. 2. That both sodium chloride and sea salt aerosol adopt a single crystal cubic morphology.

Both of these assumptions are contradicted in the literature. With respect to the first assumption, a number of infrared studies on diffusion-dried NaCl and sea-salt aerosols (Cziczo et al. JGR 102, 18843, 1997; Cziczo and Abbatt, J. Phys. Chem A 104, 2038, 2000; Weis and Ewing JGR 104, 21275, 1999; J. Phys. Chem. 103, 486, 1999) generated using the same atomizer (TSI 3076) used in this study revealed that

ACPD
7, S5169–S5172, 2007

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it was extremely difficult to prepare completely dried salt aerosol. These studies also suggested that there were significant amounts of water trapped inside the salt matrix. With respect to the assumption of cubic morphology, a number of micrographic studies (Cheng, in Atmospheric aerosols and nucleation, Wagner and Vali, eds., 1988; Cheng et al., Atmos. Res. 22, 15, 1988) have shown that salt aerosol particles tend to form extended heterogeneous structures upon crystallization rather than simple cubes. In the present study, the agreement between shape factors for NaCl and sea-salt aerosol only support the assertion that NaCl and sea-salt particles have the same morphology, not that this morphology is cubic. If the diffusion dryer is not efficient enough, the particles may in fact enter the LACIS as supersaturated droplets rather than dry particles. At the very least, the authors need to address this point and reassess the uncertainties in their measurements based on the possibility that there are significant uncertainties in the shape factor used to determine the dry particle diameter. This may significantly increase the uncertainty in the parameterized equation.

Specific comments:

p. 3: The authors have not explained the mathematical method that was used to assign the estimated sea salt composition reported in Table 2.

p. 4, equation 1: How was the solute volume (Vs) determined? Presumably this was derived from the dry mass equivalent diameter.

Figure 2/humidity measurements: It was not entirely clear to me how humidity in the LACIS was determined. As I understood it, the authors used the hygroscopic growth of particles of (NH4)2SO4 to calibrate the humidity scale. If this calibration relies on accurate knowledge of the mass equivalent diameter as well, then there may be significant uncertainty in the humidity calibration. Stylistically, when showing a 1:1 linear relationship in a calibration, it would be appropriate to use a figure with a 1:1 aspect ratio. In future, the authors might want to consider an in-situ determination of humidity using IR spectroscopy through the LACIS unless the humidity is not uniform in the flow

ACPD

7, S5169–S5172, 2007

tube due to the sheath gas.

Figures 5 and 6. The meanings of the various fits and vertical lines are not explained in the figure captions.

Figure 7. The solid line in this figure is not explained in the legend or figure caption.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 11511, 2007.

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7, S5169–S5172, 2007