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Atmospheric
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Interactive comment on “Laboratory measurements of the optical properties of sea salt aerosol” *by* R. Irshad et al.

R. Irshad et al.

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In response to editorial comments made by A. Lambert on 31 March 2008:

-1- The relative humidity within the aerosol cell and the conditioning vessel were measured directly using Honeywell RH and temperature sensors. RH was used as it is the relative humidity that determines the state of the aerosol particle in the atmosphere. The aerosol solute concentration was not known exactly and could not easily be measured. Additionally, the usefulness of the aerosol solute concentration over the RH in atmospheric analyses involving refractive index data is uncertain. However, the molality of the particles was estimated using measured size values and estimated dry sizes for the aerosol particles. This estimated molality is now given alongside RH in the revised paper.

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-2- The phrase "As the aerosol becomes wetter" has been changed to "as the water content of the aerosol increases" in the revised paper. The phrase "aerosol solution" was deliberately omitted. This is because one of the important distinctions between these measurements and preceding measurements is that they are made on actual aerosols and not on bulk solutions. The phrase "the aerosol becomes more dilute" was then also considered inappropriate due to the possible confusion between dilution of salt in water and dilution of aerosol particles in a greater volume of air. It is hoped that the phrase chosen for the final revision is an adequate compromise.

-3- Figure 5 gives values of refractive index at infinite frequency or zero wavelength (Minf) and this is estimated at 1.4 for water. However, the text that states the refractive index value of water as 1.33 refers to the refractive index at visible wavelengths. It is clear that this can cause confusion, but will no longer be an issue in the revised paper as the retrieval method was found to use the refractive index at 500nm as its fixed index reference value, rather than the refractive index at infinite frequency. This has been rectified in the revised paper.

-4- Upon analysis it was found that the real and imaginary parts of the refractive indices measured by Volz (1972) do not display the Kramers-Kronig relationship. This has been stated more clearly in the revised text.

-5- This paragraph reiterates the origin of data used for the volume mixing rule calculations performed by Shettle and Fenn (1979), as well as summarising the findings presented in the paper. To reduce the possibility of confusion, this paragraph has been rewritten and the aerosols referred to are detailed at each stage.

-6- The references suggested in the Saint-Gobain product sheet refer to a refractive index of approx. 1.89 at 0.185 microns. We give a value of approx. 2.19 for the refractive index at zero wavelength. This is derived from the relationship: dielectric permittivity at infinite frequency (E-inf) = refractive index at infinite frequency (Minf). Minf is used as a fixed index reference point for the retrieval (Bertie et al. (1994)).

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While the Minf value is difficult to measure or calculate, values of E-inf have been reported in the literature, most recently in Peyman et al. (2007) where all previous values are also summarised and compared. The value of E-inf is given as approx. 5.0 (+/-0.6), which implies a value of approx. 2.24 (+/-0.14) for Minf. Therefore the value of 2.19 obtained for dry sea salt by our model is not unduly high.

However, further analysis of the retrieval code revealed that the actual value used in the model is the refractive index at 500nm (20,000 cm-1) rather than the refractive index at infinite frequency. This value is well known both for NaCl and water, so this effectively negates the original issues due to high refractive index values. The revised paper has been corrected to refer to the refractive index values at 500nm rather than Minf.

Regarding the other references suggested, these have all previously been considered but were not specifically cited in the original paper for a number of reasons. However, they are now referenced and discussed in the revised paper. Specifically, regarding Querry et al. (1976), measurements on bulk solutions of NaCl are made at two different concentrations. As stated in the paper, this may be useful for studies of ponds and lakes but may not be directly applicable to aerosols. Additionally, no reference or comparison is made to volume mixing rules. Tang (1997) refers to mixed salt solutions of NaCl, (NH4)SO4 and water, which are not directly representative of sea salt aerosol. Also, measurements are made using the single particle levitation technique that deals with particles much larger than accumulation mode aerosol, effectively resulting in a bulk solution. The refractive index is derived only at a single wavelength determined by the He-Ne laser used for the experiment, rather than over a range of wavelengths. Similarly, Tang et al. (1997) also use the single particle levitation technique. Winter and Chylek (1997) calculate the contribution of sea salt aerosol to the planetary clear-sky albedo using data from, among others, Shettle and Fenn (1979) as well as the Bruggeman mixing rule (Chylek et al. (1988)). However, no verification of the mixing rules is undertaken, and comparisons of measured and calculated optical depths show a slight discrepancy. Yunus (1992) measures refractive index of the major component

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solutions of seawater at a single wavelength, again determined by the wavelength of a He-Ne laser. He also looks at the absorbance of the component solutions over a narrow wavelength range (200 to 1100nm) and compares them with measurements made on both artificial and natural seawater. Not only are these measurements not directly applicable to sea salt aerosol, but the paper itself concludes that the values of refractive index obtained for the components do not agree with the values of refractive index for either artificial or natural seawater. Finally Neto et al. (2004) measure the absolute refractive index of NaCl solutions, but again only at a single wavelength. While the information offered in these papers is useful, we nevertheless feel that the scope remains for further direct measurements of the refractive index of SSA over a range of wavenumbers and relative humidities.

Many thanks to Alyn Lambert for advice given and helpful comments.

References:

Bertie et al., Infrared intensities of liquids XVI. Accurate determination of molecular band intensities from infrared refractive index and dielectric constant spectra, J. Mol. Struct., 324, 157-176, 1994.

Chylek et al., Scattering of electromagnetic waves by composite spherical particles: experiment and effective medium approximations, Appl. Optics, 27, 2396-2404, 1988.

Neto et al., Determination of absolute values of refractive index of liquids using an interferometric method, Revista de Fisica Aplicada e Instrumentacao, 17, 2, 74-79, 2004.

Peymann et al., Complex permittivity of sodium chloride solutions at microwave frequencies, Bioelectromagnetic, 28, 264-274, 2007.

Querry et al., Complex refractive index in the infrared for NaCl, NaNO3 and NaHCO3 in water, J. Opt. Soc. Am., 66, 830-836, 1976.

Shettle, E. P. and Fenn, R. W., Models for the aerosols of the lower atmosphere and the effects of humidity variations on their optical properties, AFGL-TR-79-0214, 1979.

Tang, I. N., Thermodynamic and optical properties of mixed-salt aerosols of atmospheric importance., J. Geophys. Res., 102, 1883-, 1997.

Tang et al., Thermodynamic and optical properties of sea salt aerosols, J. Geophys. Res., 102, 23269-23275, 1997.

Volz, F. E., Infrared refractive index of atmospheric aerosol substances, Appl. Optics, 11, 755-759, 1972.

Winter, B. and Chylek, P., Contribution of sea salt aerosol to the planetary clear-sky albedo, Tellus, 49B, 72-79, 1997.

Yunus, W. M. M., Temperature dependence of refractive index and absorption of NaCl, MgCl2, and NaSO4 solutions as major components in natural seawater, Appl. Optics, 31, 2963-2964, 1992.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 71, 2008.