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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Radiative consequences of low-temperature infrared refractive indices for supercooled water clouds" *by* P. M. Rowe et al.

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Reviewer 2 has commented: "The authors pointed out a large difference in the Zasetsky CRI at 273 K and the Hale and Querry data at 300 K. However, they did not elaborate on the need to resolve this, nor make any suggestions on which may be more accurate. [Certainly there would be no "ice like domains" in at 273, so it would seem that the Zasetsky and Hale&Querry results should be the same at 273 or 300 K.]"

I agree with the reviewer that some elaboration on this subject is required. However, I would argue that the complex refractive indices (CRI) of water at 273 K are different from those at 300 K. A temperature-induced shift in mid-infrared spectra of water

above the freezing point has been well documented [Zasetsky et al., 2004]. "Ice-like domains" are not ice, but ordered lower-density local environments. Molecular dynamics calculations, infrared measurements, and neutron scattering experiments all indicate that these domains are present at temperatures well above 273 K (see Zasetsky et al. [2004] and references cited therein). As the temperature increases, e.g., from 273 to 300 K, the volume fraction of these domains decreases from 0.1 to 0.02 and the librational mode at 600 cm-1 shifts to lower wavenumbers. Admittedly, the absolute magnitude of this shift decreases when the temperature approaches 300 K. Nevertheless, this temperature dependence needs to be taken into account if one wishes to obtain accurate data from spectral measurements.

Reference cited:

Zasetsky, A. Y., A. F. Khalizov, and J. J. Sloan (2004), Local order and dynamics in supercooled water: A study by IR spectroscopy and molecular dynamic simulations, J. Chem. Phys., 121(14), 6941-6947.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 18749, 2013.