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Comments on "A limited-area-model case study of the effects of sub-grid scale variations in relative humidity and cloud upon the direct radiative forcing of sulfate aerosol"

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Haywood et al. [1997] (hereafter refered to as HRD) have suggested that sub-grid variations in relative humidity (RH) can, through the nonlinearity in the water uptake by aerosol particles, yield a substantially larger estimate of direct radiative forcing by sulfate aerosols than general circulation model (GCM) estimates based on the mean temperature and humidity within each grid cell. This suggestion is based on calculations of water uptake and radiative forcing using a prescribed uniform dry aerosol size distribution, with RH taken either from aircraft observations of shallow convection or from a numerical model simulation of shallow convection. In all cases considered the RH ranged from a minimum of 70–85% to a maximum of 100%, with at least 10% of the frequency at or near 100% RH.

We suggest that the direct radiative forcing due to sub-grid variations in RH is overestimated by HRD, and that treating sub-grid variations in RH may not be necessary to estimate direct radiative forcing. The largest contribution to the radiative forcing estimated by HRD comes from the fraction of the domain with 100% RH, where clouds form. HRD did not account for the fact that many if not most of the aerosol particles in the size range they considered are activated to form cloud drops in clouds, leaving only relatively small or insoluble particles within cloud as interstitial particles. If the contribution of the activated aerosol particles to the direct radiative forcing is neglected then the direct radiative forcing from the cloudy fraction is greatly reduced. Under some conditions the estimated direct radiative forcing is actually less when sub-grid variations in RH are correctly accounted for than when the grid cell mean humidity is used.

To see this, we have adopted the ammonium sulfate aerosol size distribution used by HRD, used Kohler theory [*Pruppacher and Klett*, 1997] and HRD's linear treatment of deliquescence to calculate water uptake for each of 100 particle size bins, used volume mixing to determine the wet particle refractive index, used a Mie code [*Wiscombe*, 1979] to calculate the radiative scattering for each size bin, and then integrated over the aerosol size distribution to determine the optical depth as a function of RH. Figure 1 shows the 0.55 μm wavelength aerosol optical depth as a function of RH, assuming none of the aerosol particles are activated at any RH. The optical depth at 100% RH is 13.7, far larger than the optical depth under dry conditions, 0.1. To determine the grid cell mean optical depth we integrate the optical depth over RH using the RH frequency distribution simulated by the HRD model and shown in their Figure 2a. We use 20 RH bins with width 1% and centered on RH = 80.5%, 81.5%, ..., 99.5% and an additional bin at RH=100%. The frequency for the 99.5% RH bin in HRD's Figure 2a (11.7%) includes both points with 99% <RH< 100% and points with RH $\geq$ 100%. We estimate the frequency for 99% <RH< 100% by extrapolation of the frequencies at the adjacent lower RH bins. This gives a frequency of 5.1% for 99% <RH< 100% and 6.6% for RH$\geq$ 100%. Integrating the optical depth using the RH frequency distribution simulated by the HRD model, the mean optical depth is 1.71, while the optical depth at the mean RH (93.6%) is 0.62. The ratio of the two estimates, 2.73, is significantly larger than the ratio of radiative forcing calculated by HRD (1.73) because the optical depths at high RH are large enough that the radiative forcing is not proportional to the optical depth. The ratio of the aerosol layer reflectance using a two-stream delta-Eddington model [*Joseph et al.*, 1976] and a solar zenith angle of 53° is 1.74, in excellent agreement with the ratio of radiative forcing calculated by HRD. (Thus, if HRD had used a much smaller aerosol concentration or a thinner aerosol layer, their estimate of the ratio of radiative forcings would have been much closer to 2.73 than 1.73.)

Having demonstrated consistency between our method and the HRD method of calculating the impact of subgrid scale variations in RH on radiative forcing, we can now consider the impact of accounting for the influence of aerosol activation in the cloudy fraction of the grid cell. As an extreme case, if we assume that all aerosols are activated in the cloudy fraction then the grid cell mean optical depth of the remaining aerosol is 0.81, only 30% larger than the optical depth at the mean RH, 0.62. If we assume that only aerosol particles with dry radius larger than 0.1 μm are activated (which is predicted by the aerosol activation parameterization of *Ghan et al.* [1993] for an aerosol number concentration of 6000 cm^{-3} and an updraft velocity of 1.4 m s^{-1}) then the grid cell mean optical depth is 0.97, which is 56% larger than the optical depth at the mean RH but much smaller than the mean optical depth calculated assuming none of the aerosol particles are activated.

The treatment of aerosol activation matters even more if the cloud fraction is somewhat higher. For example, if the RH frequency distribution is represented by a normal distribution with a median RH of 100% and a standard deviation of 5% so that the cloud fraction is 50%, then for the same dry aerosol the grid cell mean optical depth is 0.63 assuming

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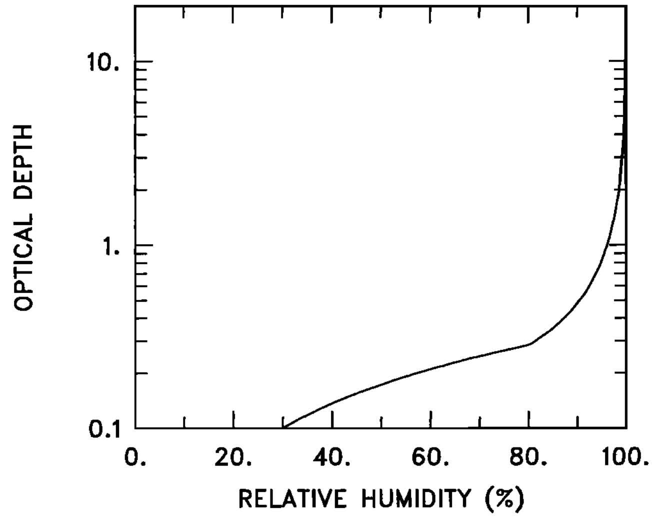

Figure 1. Aerosol optical depth at at wavelenght of 0.55 μm plotted as a function of relative humidity for an ammonium sulfate aerosol with a log-normal size distribution (a mean radius of 0.05 μm and a geometric standard deviation of 2) and a column number concentration of 3×10^8 cm^{-2}.

all aerosol particles within cloud are activated, 1.93 assuming only particles larger than 0.1 μm radius are activated, and 7.6 assuming none of the particles are activated. The optical depth at the grid cell mean RH (98.0%) is 1.50.

To summarize, HRD suggested that, because GCMs do not account for sub-grid scale variability in RH and its impact on water uptake and aerosol radiative properties, GCM estimates of direct radiative forcing are systematically too low. We suggest that, because water uptake is dominated by conditions that favor aerosol activation and produce cloud, the GCM estimates of direct radiative forcing may not be so far off. Direct radiative forcing within sub-grid scale clouds is likely to be negligible because so many of the aerosol particles in clouds are activated, and hence cannot contribute to direct radiative forcing (although activated aerosol material in cloud droplets still scatters sunlight, the effective radius of the cloud droplets is much larger than that of the sulfate particles, so that the scattering by sunlight is much less than if the particles remained suspended aerosols). Correspondingly, however, there is the potential for indirect radiative forcing by these sub-grid scale clouds, and a GCM neglecting sub-grid scale clouds would neglect this contribution to indirect radiative forcing. The calculation of this indirect radiative forcing involves consideration of competition between natural and anthropogenic aerosol particles as cloud condensation nuclei and is beyond the scope of our comments. The reader should also note that, because water uptake and aerosol activation are closely related processes, these conclusions depend little on the aerosol particle composition; although insoluble particles are less likely to be activated, water uptake is much less than for the more readily activated soluble particles. Thus, the conditions most likely to produce the greatest water uptake (high RH and large soluble aerosol particles) are also those conditions most favorable for aerosol activation.

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References

Ghan, S. J., Chuang, C. C., and Penner, J. E., A parameterization of cloud droplet nucleation. Part I: Single aerosol type, *Atmospheric Res., 30,* 197–221, 1993.

Haywood, J.M., Ramaswamy, V., and Donner, L.J., A limited-area-model case study of the effects of sub-grid scale variations in relative humidity and cloud upon the direct radiative forcing sulfate aerosol, *Geophys. Res. Lett.,* , *24,* 143–146, 1997.

Joseph, J.H., Wiscombe, W.J., and Weinman, J.A., The delta-Eddington approximation for radiative transfer, *J. Atmos. Sci., 33,* 2452–2459, 1976.

Pruppacher, H.R., and Klett, J.D., *Microphysics of Clouds and Precipitation.* Kluwer, Dordrecht, 954 pp., 1997.

Wiscombe, W., Mie scattering calculations: Advances in technique and fast, vector-speed computer codes, *NCAR Technical Note NCAR/TN-140+STR,* available from National Technical Information Service as NTIS PB 301388, 1979.

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