**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "A redistribution of water due to pileus cloud formation near the tropopause" *by* T. J. Garrett et al.

T. J. Garrett et al.

Received and published: 8 February 2006

We greatly appreciate the attention that the editor has drawn to this problem, particularly as it appears that we were indeed mistaken as to the appropriate time scale to apply.

Looking at the issue more closely, we believe that there are in fact three different time scales of potential relevance to haze droplet growth. Applying dimensional analysis, the first is

$$\tau_1 \sim r^2/D$$

where D is the diffusivity and r the droplet radius. τ_1 represents the approximate time required to create a steady-state vapor field around the droplet under a change in environmental conditions. This time scale is very short, of order microseconds at most. We

Full Screen / Esc

Print Version

Interactive Discussion

Discussion Paper

EGU

have assumed here an effective diffusivity for D with a value for the accomodation co-efficient α_c of 0.036, although there is some contention regarding the true value of α_c, some assuming a value close to unity, and others finding lowered values due to impurities (Pruppacher and Klett, 1997; Karcher and Koop, 2005). While τ_1 is the timescale we mentioned in the paper, it perhaps should have been obvious that a different time scale would be of more relevance, as nowhere does τ_1 include a parameter (such as liquid density) relevant to the liquid phase, and hence aerosol activity.

The second time scale is the one the editor highlighted

$$\tau_2 \simeq 1/\left(4\pi r D N\right)$$

where, N is the droplet concentration. This time scale is particularly useful for estimating the time required to "quench" excess saturation following nucleation of haze droplets. In other words, it is the time required to deplete the ambient relative humidity to the equilibrium vapor density over the surface of the droplet. For the case we consider in our manuscript, with haze particles rarely exceeding $r = 0.1\ \mu$m, this time scale is of order $\tau_2 > 1000$ s, which is very long. Note that the radius is in the denominator here - small particles are inefficient at sucking out vapor from the atmosphere.

The third time scale τ_3, which perhaps should have been the one discussed, is the time required for a haze aerosol to accumulate sufficient water mass for its activity a_w to adjust to the environmental relative humidity. The activity of a solution droplet is a function of its radius alone. Thus, the basic equation required to calculate $a_w\left(t\right)$, derived from Maxwellian growth considerations, is

$$r\frac{dr}{dt} = \frac{D\Delta\rho}{\rho_w}$$

where, ρ_w is the water density, and $\Delta\rho$ is the vapor density difference between the surface of the droplet and the environment. Latent heat considerations are ignored

here because we are concerned with cold temperatures near 200 K and low water contents. Scale analysis dictates that

$$\tau_3 \sim \frac{\rho_w r^2}{\rho_s D}$$

where, ρ_s is the saturation vapor density. Here, radius is in the numerator: the activity of small droplets adjusts quickly to environmental vapor density changes because the mass accumulation required for full equilibration is small. An opposing effect is that the flux of molecules to the droplet surface is slowed in an environment with low vapor concentrations (as estimated by ρ_s). For typical haze droplet sizes and the temperatures that we considered in our paper, $\rho_s \sim 1 \times 10^{-6}$ kg m^{-3}, $r \sim 0.1$ μm, and thus $\tau_3 \sim$10 s.

So certainly, it appears we were incorrect in our statement that the appropriate time scale is τ_1. However, τ_2 is probably not right either, because for haze aerosol it is simply too long. Rather, equilibrium between the haze aerosol and its environment is reached most quickly through τ_3. At the relatively rapid cooling rates of 0.03 K s^{-1} we considered, the timescale for changes in ρ_s is ~ 100 s. Thus, adjustment of a_w under very rapid cooling will be a concern for aerosol with sizes near 1 μm. However, since we consider mostly aerosol that were much smaller, τ_3 represents a sufficiently short lag time that we expect it to be of negligible importance to our results.

In any case, in our simulations, the ultimate sizes of pileus ice crystals were dictated much less by the aerosol activity than by the concentrations of aerosol, all of which were easily nucleated.

To accomodate the editor's concerns, we have removed the text referencing KarcherKoop2005.

References

Kärcher, B. and Koop, T.: The role of organic aerosols in homogeneous ice formation, Atmos. Chem. Phys., 5, 703–714, 2005.

Pruppacher, H. R. and Klett, J. D.: Microphysics of Clouds and Precipitation, 2nd Rev. Edn., Kluwer Academic Publishing, Dordrecht, 1997.

Full Screen / Esc

Print Version

Interactive Discussion

Discussion Paper

EGU