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

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A revised paper "Convective formation of pileus cloud near the tropopause" by Garrett et al. has been send out for review. A final decision will be made by the Editor after all reports have been received. In view of the holidays to come, an extended time for final review comments will be allowed.

Let me reiterate one issue that came up during review of the original manuscript as published in ACPD. In their reply to the Specific Comment 5 raised by Reviewer 1 (large supercooled aerosol particles may get out of equilibrium with ambient water vapor), Garrett et al. state that ".. the time scales associated with diffusion of water vapor to the surface of an aerosol are simply far too short not to adjust to ambient

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atmospheric cooling." In fact, in the revised version, Appendix A2 still contains this argument, specifying the above time scales as being of the order r^2/D, with the particle radius r and the (effective) diffusivity D.

As Garrett et al. refer to work published by Kärcher and Koop (2005), I will comment on this issue in addition to reviewer 1 (who gave a wrong citation, Haag et al. (2003) would be the correct one I presume) and expect Garrett et al. to consider my argument.

I suspect that Garrett et al. confuse two time scales that have distinct physical meanings. There is the time scale r^2/D mentioned by Garrett et al. in their revision. In contrast to their statement, this is the time required for diffusion of water vapor to attain a steady-state concentration profile in air toward the particle surface. Garrett et al. are correct that this time scale is very short (milliseconds or less of submicrometer particles). It is exactly this observation that permits steady-state diffusion theory to be applied in calculating vapor transport by condensation (Pruppacher and Klett, 1997).

The time scale on which Garrett et al. should probably base their argument is $1/(4\pi r D n)$, with n being the concentration of particles of size r toward the moelcules condense. Inspection of this time scale shows that it can take seconds to minutes until water vapor is completely transported to few large particles at TTL temperatures. This time scale is derived in many textbooks (e.g., Seinfeld and Pandis, 1998). I suspect, however, that the issue of possible non-equilibrium water uptake is not crucial for the work of Garrett et al. in which case the authors should revisit their new Appendix A2 and shorten the text accordingly.

Finally I like to clarify that Kärcher and Koop (2005) distinguish between two processes affecting the freezing behavior of organic-rich liquid aerosol partucles. The first is concerned with a reduced hygroscopicity of many particulate organic matter versus sulfate, which directly affects the freezing particle volume. The second point is the non-equilibrium effect discussed above. For typical aerosol particle sizes (100 nm), the non-equilibrium effect is negligible, unless the accomodation coefficient may become

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very small in case of organic aerosol composition effects on surface kinetics.

References

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Interactive comment on Atmos. Chem. Phys. Discuss., 5, 8209, 2005.

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