

Interactive comment on “A parameterisation for the activation of cloud drops including the effects of semi-volatile organics” by P. J. Connolly et al.

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This manuscript presents a parametrisation of the effect of semivolatile organics on cloud drop activation. Although the parametrisation is based on a modeled mix of oxidation products of volatile organics, and therefore necessarily contains uncertainty, I believe that it is a useful first step toward regional and global modeling of the effect. However, I have major concerns that should be addressed before I can recommend acceptance to ACP. These come largely from my experience on studying the effect of nitric acid on cloud drop activation. Nitric acid should behave quite similarly as the organics with similar volatility and effective Henry's law coefficient (the main difference being that nitric acid is a smaller molecule and hence diffuses faster), so I believe that

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lessons learned with nitric acid are relevant. The present parametrisation appears to reproduce the parcel model simulation results very well, but the simulations have only been carried out for a limited range of conditions. Also, I would like to be convinced that the success of the parametrisation is not due to cancellation of errors related to including both very low volatility species and very high volatility species which behave very differently at cloud activation but are lumped together in the parametrisation.

The parametrisation is based on the assumption that all of the gas-phased organics partition to the aerosol phase at cloud base (BTW, does this mean the location of 100% RH or that of the maximum supersaturation?). I am quite surprised by this, as I know that in many cases nitric acid does not partition completely to the growing droplets before S_{max} is reached, and the organics in the two or three most volatile bins of the volatility distribution (see Topping et al, Nature Geoscience 2013) are more volatile and have smaller Henry's law constants than nitric acid. The partitioning between the gas and the droplets at S_{max} depends on updraft velocity, temperature and total pressure (as T and P affect the diffusion velocities of both water vapour and the co-condensing species) and available droplet surface area. The authors justify the assumption of complete partitioning to droplets by the calculation shown in Topping et al (Fig. 2 there), but that was done for a size distribution with a very large median diameter, 150nm, while the median diameter in the present manuscript is only 60nm. I strongly suspect that the partitioning to the droplet phase is clearly less for the 60nm size distribution regardless of the number concentration. Careful reading of Fig 1 a) of the present paper seems to corroborate this: for example, with the aerosol concentration of 500 per cc, something like 15% of the organics remains in the vapor phase at cloud base. (And as I explain below, I think that about one third of all organics – the least volatile species – should be left out of the simulations altogether, in which case the percentage remaining in the gas phase becomes even higher.)

Secondly, I am not completely convinced by the justification given for the constant shift of the size distribution (i.e. constant arithmetic SD) due to condensation of the organics.

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Fig. 2 shows what happens at an RH of 95% where only the most involatile organics condense (species which have clearly lower volatility than nitric acid). I would like to see the shift of the size distribution at cloud base.

Regarding the low volatility organics with $\log_{10}(C^*)$ between -6 and -2 (or -1). As can be seen from Figs. 1 a) and b), these substances condense rapidly (about half of the organics are depleted within a few tens of seconds when there is sufficient aerosol surface area present) at RH's clearly below 100%. In fact, these low volatility substances are capable of condensing at RH's much less than 95%, and I believe that in reality (and also in large scale models having advanced partitioning or condensation schemes for organics), it is very rare that they remain in the gas phase when RH has increased to 95% (or even 90% for that matter) as is assumed in the present work. This is because the condensable organics are produced photochemically at sunny conditions when there is sufficient OH to oxidize the less volatile species, and by the time the RH has increased to 95%, the conditions are usually pretty cloudy. I therefore think that these low volatility species should not be included in the parcel model simulations at all.

I also think that the comparisons between the parcel model simulations should be done for a broader range of conditions than shown in the manuscript in order to make a convincing case of the goodness of the parametrisation. The median diameter and the SD of the aerosol size distribution should be varied sufficiently, and at least some of the comparisons should be repeated at a lower temperature. It would also be interesting to know what the effect of possible insoluble core material in the aerosol is. The authors should make plots from parcel model runs that clearly show what fractions of the three most volatile bins condense before cloud base at different temperatures and updraft velocities, and with different aerosol size distributions. If important effects are seen, these effects should be included in the parametrisation.

As noted above, the nitric acid effect on cloud activation is very similar to that produced by semivolatile organics. The literature should therefore be cited (especially

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papers in which parcel model simulations have been carried out). Furthermore two recent papers (Makkonen et al., ACP 12, 7625, and Xu and Penner, ACP 12, 9479) examined the global radiative effect of nitric acid via enhanced cloud activation. The effect simulated by Makkonen et al. was about twice as big as that found by Xu and Penner. The reason for the discrepancy is unclear, but it may have to do with the different parametrisations. It appears that the present parametrisation is at least somewhat similar to that used by Xu and Penner, and it would be interesting to see discussion about the similarities/differences of the two parametrisations.

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