

## Review of “Can Semi-Volatile Organic Aerosols Lead to Less Cloud Particles?”

This paper investigates the sensitivity of cloud droplet activation, using the Abdul-Razzak et al. (2000) parameterisation, to the aerosol chemical composition, mass and number concentrations, and particle size. The main finding is that the simulations suggest that semi-volatile compounds almost always lead to fewer cloud droplets than without semi-volatiles. Whilst I agree this area is worthy of further investigations, I believe the main finding is an artefact of the parameterisation used – the Abdul-Razzak et al. (2000) scheme and perhaps also in the way it is implemented in the model

There are a few statements that lead me to conclude the authors might not be treating activation in the correct way. For example, on line 141 the authors write “the activated number concentration is only a function of number concentration and dry particle diameter”. This is not true: in addition to environmental parameters such as temperature and pressure, activation is also a function of the geometric standard deviation (see equation 15 of ARG, 2000), and the aerosol chemistry (see equation 3 of ARG, 2000).

Of the parameters above the geometric standard deviation is an important parameter for cloud drop activation. If the distribution is narrow (small geometric standard deviation) then the competition effect will be small and more particles will activate at once. If the aerosol size distribution is broad / geometric standard deviation is large one tends to find that fewer particles activate. Connolly et al. (2014) showed that for single aerosol modes it was necessary to shift the geometric standard deviation to smaller values in order to predict activated fractions more accurately – see their equation 15. Crooks et al. (2018) have now extended this result to multiple modes.

Please clarify whether this shifting done in the current manuscript.

On line 152 the authors mention that, in the Abdul-Razzak et al. (2000) scheme, increasing number concentration decreases the ambient supersaturation, which reduces the number activated, therefore suppressing activation. This argument is slightly circular though since to reduce the ambient supersaturation more particles must have been activated.

We know that the Abdul-Razzak et al. parameterisation does not always predict the correct response to inputs. Connolly et al. (2014b) showed a comparison between the Abdul-Razzak et al. (2000) and the Fountoukis and Nenes (2005) parameterisations. Their Figure 3(a-d) is reproduced in Figure 1, below. Figure 1a shows how increasing the total aerosol mass (by increasing the aerosol particle number concentration, the x-axis) eventually leads to less particles being activated in the Abdul-Razzak et al. parameterisation. Such a reduction is not seen in the Fountoukis and Nenes (2005) parameterisation (see Figure 1d).

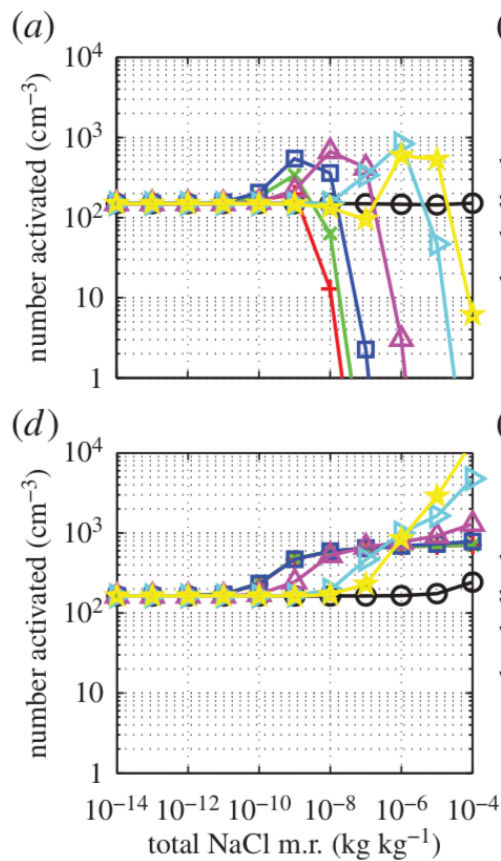


Figure 1. shows the activated fraction when adding NaCl particles to the aerosol population with a total mass loading indicated by the x-axis. Colours refer to different modal diameters of the NaCl particle size distribution (see Connolly et al, 2014b for full details). (a) is for the Abdul-Razzak et al. parameterisation; (b) is for the Fountoukis and Nenes parameterisation.

The results in the presented manuscript, that fewer particles are activated with semi-volatiles for higher updrafts, are also in contrast to Connolly et al. (2014), which found ARG at low updraft speeds activated fewer particles with semi-volatiles switched on; (see Figure 6 of Connolly et al, 2014). I suspect the reason for this contrast between the two studies is that the Abdul-Razzak et al. parameterisation gives a different response when multiple aerosol modes are used, as has been shown by Connolly et al. (2014b). Indeed results by Simpson et al (2014), which are reproduced in Figure 2, indicate that this is the case. The ARG parameterisation results are indicated by the '+' symbols and it is shown that ARG is further below the 1 : 1 line when the updrafts are high (red) vs when the updraft is low (blue).

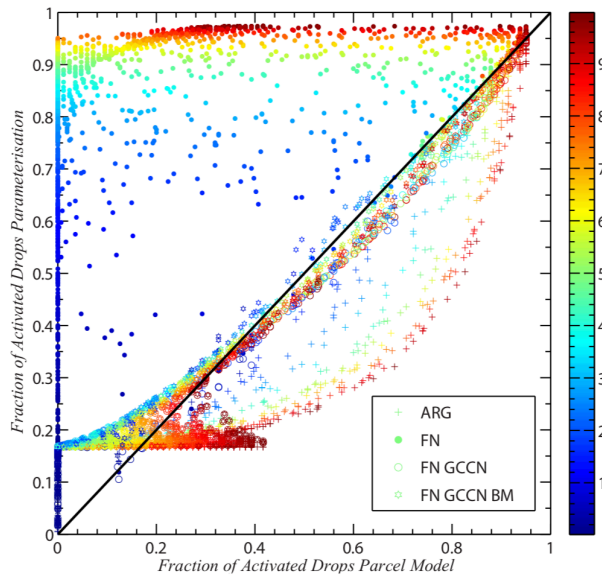


Figure 2. Results from Simpson et al. (2014). Using a bimodal aerosol size distribution. Symbols coloured by updraft velocity (m s<sup>-1</sup>).

On line 126 there is mention of simulations looking at activation at extremely low humidity. How relevant are these simulations, given that activation would not occur at low RH anyway?

The statement on line 188 about the discrepancy between the results presented and those of Topping et al. (2013) seems to indicate that the differences are because Topping et al. (2013) resolved more physics than the cloud drop activation process. In fact this is not really true. The Topping et al. study only considered condensation until the point of cloud drop activation.

Unfortunately, because of these shortcomings I feel like the conclusions drawn about most areas on earth experiencing less CCN that currently thought, except the more polluted & dry areas, are all dependent on the parameterisation used and its implementation.

## References

Connolly, P. J., Topping, D. O., Malavelle, F., and McFiggans, G. 2014. A parameterisation for the activation of cloud drops including the effects of semi-volatile organics, *Atmos. Chem. Phys.*, 14, 2289-2302, <https://doi.org/10.5194/acp-14-2289-2014>

Connolly PJ, McFiggans GB, Wood R, Tsiamis A. 2014b Factors determining the most efficient spray distribution for marine cloud brightening. *Phil. Trans. R. Soc. A* 372: 20140056. <http://dx.doi.org/10.1098/rsta.2014.0056>

Simpson, E., Connolly, P., and McFiggans, G., 2014. An investigation into the performance of four cloud droplet activation parameterisations, *Geosci. Model Dev.*, 7, 1535-1542, <https://doi.org/10.5194/gmd-7-1535-2014>