

***Interactive comment on* “The relationship between aerosol and cloud drop number concentrations in a global aerosol microphysics model” by K. J. Pringle et al.**

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We appreciate the helpful comments of the referee. The issues raised are well taken and we have responded to every comment made by the referee.

1. *P3210 L4: The recent mechanistic parameterizations should be cited.*

Added the references: Abdul-Razzak and Ghan (2000); Nenes and Seinfeld (2003); Fountoukis and Nenes (2005)

2. *P3212 L4-5: Which system is the binary homogeneous nucleation for? Is it based on theory, laboratory, or field measurements? My understanding is that the picture of atmospheric nucleation is rather murky now. Multiple nucleation*

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mechanisms, including some "exotic" ones (e.g., ion-mediated), exist in the literature. Is your approach sufficient enough?.

New particle formation is treated through the binary nucleation scheme of Kulmala et al. (1998), which was derived from theoretical calculations and compared to laboratory measurements. We agree with the reviewer that that other nucleation mechanisms also contribute to aerosol loading (e.g. tertiary nucleation and ion-induced nucleation), but these more "exotic" parameterisations were not considered in this simulation as they are generally still under-development.

To clarify, we have added the following text:

"The binary homogeneous nucleation scheme used is that of Kulmala et al. (1998). Other nucleation processes contribute to particle formation in the troposphere, thus the model may underestimate particle number, particularly at smaller sizes and in continental regions (see Spracklen et al., 2006). To account for sub-grid nucleation, 2% of SO₂ emissions are assumed to emitted in particulate form (Spracklen et al., 2005; Adams and Seinfeld, 2003)."

- P3212 L19-20: It is entirely possible that you got right answers for wrong reasons. For example, organic carbonaceous aerosols are a very important, if no dominant, source of CCN in the stratocumulus region off the west coast of Africa, of course during the biomass burning season. This has to be properly acknowledged.*

The effect of including BC/OC emissions on the aerosol distribution in GLOMAP has been examined by (Spracklen et al., 2007), who show marine aerosol size distributions with and without the additional emissions. They find that the aerosol

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number loading is increased by the additional emissions, but they also find that it has a relatively small effect on the aerosol size distribution; changing the mean diameter of the Aitken / accumulation modes in only a few regions.

We have changed from (P3212 L19-20):

“The model treats sulfate and sea-salt aerosol only. We recognise that there are many additional aerosol species that contribute to aerosol loading, especially in continental regions. The lack of other aerosol species in our study is a limitation, but in a detailed comparison with observations, Spracklen et al. (2007) showed the model to simulate realistic distributions of aerosol number and size in marine regions (where this paper will focus).”

Has been changed to:

“The model treats sulfate and sea-salt aerosol only. The lack of other aerosol components such as elemental and organic carbon means that the model will underestimate aerosol number in many polluted regions. However, a comparison with observations (Spracklen et al., 2007) showed that the model simulates realistic distributions of aerosol number and size in most remote marine regions (where this paper will focus), with only a small improvement in the comparison when emissions of carbonaceous particles were included.”

4. *P3212 L24-25: This statement on the relative importance of size and composition is way too loose. If a particle is entirely composed of insoluble species, it will not activate irrespective of size. I know that this is an extreme case, but it illustrates the point.*

We have changed from (P3212 L24-25):

“The chemical composition of an aerosol particle can affect the activation of the particle. This additional dimension to the aerosol activation process is not captured in these simulations. This is reasonable as a first approach as size matters more than composition for the cloud nucleating ability of aerosols (Dusek et al., 2006; Andreae and Rosenfeld, 2008).”

Changed to:

“The chemical composition of an aerosol particle can affect the activation of the particle. This additional dimension to the aerosol activation process is not captured in these simulations, as we only treat hydrophilic particles. Although neglecting the effect of particle composition is a simplification, it is reasonable as a first approach as Dusek et al. (2006) have shown that the cloud condensation nuclei concentration is mainly determined by the aerosol number size distribution, with the aerosol composition playing a secondary role. This is because the amount of soluble mass in an aerosol particle changes with the third power of particle diameter but changes only linearly with a change in the soluble mass fraction. Thus a small change in diameter has a larger effect on the amount of soluble mass (and thus the activation potential) than an equivalent change in the soluble mass fraction (see e.g. Andreae and Rosenfeld (2008)).”

5. *A general question is how long your integration is. And how often is the activation calculation?*

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We have added the additional text :

(P3213, L1): The NS03 parameterisation is called at the end of each microphysical model time step, i.e. every 7.5 minutes (Spracklen et al., 2005).

(P3212, L7): The model data presented here is for October 2001, the model was spun up for three months before this time.

6. *P3213 L22: Wrong spelling. It should be "Bennartz". Please also fix it in the reference.*

Changed.

7. *P3215 L20: It is not obvious to me at what aerosol number the "flattening"; starts at the high updraft. Could it be because some data points are above 600 per cc, thus not showing up in the plot?*

In the higher updraft plot, there are a few points above the 600 cc level, but these do not change the shape of the plot at all. Because in these simulations GLOMAP only considers sulfate and sea-salt aerosol, there are few aerosol number concentrations above 1200 cc, and it with these high concentrations that the flattening at the higher updraft becomes more important.

To clarify, we have changed:

“The dependence of CDN on aerosol number, however, is dependent on
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the updraft velocity. With the higher updraft velocity the rate of change of CDN with aerosol number is increased and the flattening of the CDN / aerosol number curve occurs at higher aerosol numbers.”

To:

“The dependence of CDN on aerosol number, however, is dependent on the updraft velocity. In the lower updraft case, the rate of change of CDN with aerosol number nears zero above moderate aerosol loadings. In the higher updraft case, the rate of change of CDN with aerosol number decreases with increasing aerosol number, but is always above zero.”

8. *P3217 L12-13: Are you assuming a fixed width for the log-normal distribution?*

Yes. The lognormal distribution has a $\sigma = 1.8$. We have clarified in the text:

“The figure shows the contoured results of several hundred parcel model calculations of CDN assuming a log-normal accumulation mode (standard deviation (σ) = 1.8) with varying size and number concentration.”

9. *P3219 L20-23: Is there any physical explanation for this? "In-cloud processing" is singled out, but no discussion on its effect.*

In-cloud processing is the main mechanism which contributes to the growth of particles in the accumulation mode in the MBL (Hoppel et al., 1986). It is shown in Figure 3 that aerosol distributions in which the accumulation mode has been grown to larger sizes generally produce larger CDN concentrations,

thus we deduce that in-cloud oxidation is the main process by which the aerosol distributions become distributions that contribute to CDN concentrations that are greater than average (for a given aerosol number concentration). We have clarified by re-wording the statement:

We change (P3921, L20) from: “Thus we find that for for the same total aerosol number (and updraft velocity), marine aerosol distributions - especially those that have undergone in-cloud processing - typically produce larger CDN concentrations.”

Changed to:

Thus we find that for the same total aerosol number (and updraft velocity), marine aerosol distributions - especially those in regions where low cloud cover is extensive - typically produce larger CDN concentrations. We conclude that this is due to the ability of the cloud decks to contribute to the growth of the mean diameter of the accumulation mode - producing aerosol particles that are more able to contribute to cloud droplet formation.

10. *P3223 L7: Again, can you offer any physical explanation? Note that both the CCN and the droplet number concentrations over the Arctic are already rather low in the base case (Figure 1). This leads me to another question. Can your model capture the well-observed seasonality of Arctic aerosols (i.e., hazy conditions in winter and early spring)? If not, are the Arctic-specific findings based on your model simulations robust enough, particularly in light the statement made in P3224 L21-22?*

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We have added an additional caveat to the Arctic conclusions:

An additional point is that all correlation relations underpredict CDN in the Arctic, sometimes by as much as 75%. Simulating the Arctic aerosol is a tough challenge for aerosol models as it is a region dominated by long range transport with a complex meteorology and relatively few observations. Korhonen et al. (2008) examined the performance of the GLOMAP model in the Arctic region and found that the model consistently under predicts aerosol loading in the Arctic and is only partially able to capture the Arctic spring haze. This has implications for the calculation of CDN in this region as the low aerosol concentrations mean that there is very little competition for the available water vapor, thus aerosol can activate down to very small sizes (e.g. Komppula et al., 2005). In the calculation of the correlation relationships, aerosol number was only counted for particles > 50 nm diameter (following Jones et al., 2001), but in regions of extremely low aerosol loading a significant fraction of the activated particles are smaller than 50 nm. Under these conditions, empirical relations derived from other locations do not work well because of the fixed minimum diameter assumed. The under-prediction of Arctic aerosol concentrations in the model will exaggerate this effect. Thus, the differences between the mechanistic and empirical calculations are likely to be an upper estimate.

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