

***Interactive comment on* “Explaining global surface aerosol number concentrations in terms of primary emissions and particle formation” by D. V. Spracklen et al.**

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We thank the 2 referees for their useful and constructive comments. We have responded to all the referee comments and made modifications to our manuscript as appropriate. Referee comments are in *italic*, our response in regular text.

Referee 1

1) *Abstract, last sentence: I found the phrase “we derive optimum rate coefficients” slightly misleading. Rather than derive, the authors simply choose the coefficients (from a rather limited set of values simulated) that give the best match globally. The truly optimum coefficients are very likely to vary from site to site, and even from day to*

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day.

We agree with the referee and reword the abstract to:

“The nucleation constants that result in best overall match between model and observed CN concentrations are consistent with values derived from detailed case studies at individual sites.”

To clarify, we also insert the following line to the abstract:

“In our model kinetic and activation-type nucleation parameterizations result in a similar agreement with observed monthly mean CN concentrations.”

2) *Mineral dust emissions are not simulated; however, they can affect the CN concentrations at least at the Southern European and possibly some of the Asian sites. This should be discussed.*

We add the following discussion to the paper:

“Primary aerosol sources can suppress nucleation through providing a condensation sink for sulfuric acid vapor and scavenging newly formed particles through increasing the coagulation sink. Previous studies have shown that dust particles can reduce CN concentrations within dusty regions by up to 20% through this mechanism [Manktelow et al., 2010]. Future studies must therefore study the contribution of dust, primary biological aerosol particles [Heald and Spracklen, 2009] and trace metal emissions [Birmili et al., 2006] to particle surface area and the corresponding suppression of new particle formation.”

3) *Are particles in the hydrophobic distribution that become hydrophilic moved the second distribution? What is the criterion for this and how sensitive are the results to the assumption used? Is SOA assumed soluble?*

To clarify our model setup we add the following line to p26283, line 21:

“We assume that a single monolayer coating of soluble material is sufficient to age

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non-hydrophilic to hydrophilic particles.”

We assume SOA to be soluble. On line 20 we now state that soluble gas-phase species are sulfuric acid and SOA precursors. We change “We add SOA to the hydrophilic aerosol distribution.” to “We assume that SOA is hydrophilic”.

We describe some additional model simulations which test the sensitivity to this assumption:

“In all the experiments described here we assumed that a single monolayer coating of soluble material is sufficient to age non-hydrophilic to hydrophilic particles. We test the sensitivity to this assumption by running one simulation where we increased the amount of soluble material required to age non-hydrophilic particles by a factor of 5. This reduces the rate of ageing and increased the global surface mean concentration of non-hydrophilic particles by a factor of 2. The resulting increase in pre-existing aerosol surface area suppresses nucleation and reduced hydrophilic particle number by 10% (in a scenario where the secondary source of particles is from binary homogeneous nucleation). Total CN concentrations (hydrophilic and non-hydrophilic particles) is relatively insensitive to this change decreasing by only 4% globally.”

4) *Although commonly used, the assumption of 2.5% of SO₂ emitted as primary sulphate is quite uncertain. Thus one other sensitivity experiment that I would have liked have seen is a smaller fraction (even 0%) emitted as primary matter. Has such option been run with the model (maybe for some other study)? How would one expect a smaller value to change the results?*

As the referee points out there is considerable uncertainty in emissions of primary sulfate. Our baseline scenario assumes that primary sulfate particles are emitted using the size distribution suggested by AEROCOM. For most of the anthropogenic sulfate emission sources this is assumed to be at a diameter of 500 nm. Emitting primary sulfate at such large sizes means the number emission of primary sulphate is small and consequently has little impact on CN number concentrations. We test the sensitivity

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of modelled CN to the number emission of primary sulfate particles by reducing the assumed diameter of sulfate particles to that used by Adams and Seinfeld (2003). Such a reduction in the size of primary sulfate increases the number emission of primary sulfate particles by more than a factor of 1000. These two sensitivity scenarios are likely to span the uncertainty range in the number emission from primary sulfate and it cover the range used by previous studies.

5) *How representative of their surroundings are the surface sites? The model grid is quite coarse and thus a comparison to observations may not be as straightforward as assumed here.*

This is an important question. We chose sites that are likely to be broadly representative of the regional conditions. We do not include any sites within urban environments due to the coarse spatial resolution of our model (see page 26388).

6) *Concerning calculation of NMB, I assume that O_i is the multiyear annual mean from the observations? How sensitive is the simulation result to the year simulated and consequently then, is NMB as defined here a good measure of the model bias?(I do understand that it is not possible to run a computationally intensive global model for the whole measurement period.)*

Correct, O_i is the observed multiyear annual mean. We clarify in this in the text.

Ideally, we would like to run the model over the entire measurement period. However, as the referee acknowledges the long time period of the observations preclude direct year to year comparison of model and observations. Because we are comparing model and observations over long time periods our choice of one model year is not likely to be very important. We add the following discussion to page 26388:

“The long sampling period (monthly and annual means) used in this analysis means that meteorological variability between specific years of observation and the year 2000 simulation is not likely to be a significant factor in the comparison.”

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7) *Figure 3: Either add the regression lines to the plot or give the regression equations somewhere else. The reader automatically tries to fit by eye the equation to the given data points, which is less than straightforward when the axes are logarithmic. If possible, remove the unnecessary plus signs from the equations in plots a, c and d. Try and plot regression lines on Fig. 3.*

We remove the regression equations from the figures as suggested.

8) *I appreciate the idea to use an additional data set to validate the model over the remote oceans. However, I would question the representativeness of the Heintzenberg data set on most latitude bands since it is based on few campaign measurements at only few locations of the latitude bands. Thus, if the model doesn't agree with this data, it doesn't necessarily mean that the fault lies in the model (whereas I think it would be safer to say so in the case of the 36 long term sites).*

We include the following sentence to inform readers about the potential lack of representation in the Heintzenberg database.

“Additionally, the limited number of field campaigns available within certain latitude bands means that the dataset may not adequately represent CN concentrations within these regions. For this reason we only make qualitative comparisons between the dataset and our simulations.”

Referee 2

1) *Now the comparisons are made between measured and modeled total "CN" concentrations, without further information on the particle size distributions - although it is well known that the size distribution has a major effect on e.g. the climatic implications of the results. The authors state, however, that for many of the studied sites size distribution data was available. It would be nice if the authors could discuss a little the potential agreement/disagreement between the modeled and measured size distributions.*

We use total particle number as this is the quantity that is most widely available and

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is strongly impacted by particle formation. We agree that an analysis of aerosol size distributions is important, but we feel that it is outwith the scope of this study. It would also limit the spatial extent of our analysis quite substantially as only 13 of the 36 sites have size distribution data (see comment below and new version of table 2). We include in a modified discussion section the importance of evaluating particle size distributions in future work.

2) *It would also be nice if the authors could give the instrument that the CN data from each studied station is based on as well as the measured size range (maybe in Table 2).*

We modify table 2 to give the type of instrument used.

3) *Although the agreement between the measured and modeled data is generally pretty good, it is not perfect (I assume particularly if the size distribution would be concerned). It would thus be good if the authors could elaborate a little on what are the next important steps in developing the aerosol representations in global models.*

This is a good suggestion. We add/modify the discussion as shown below:

“Our comparison between monthly mean model and observed total particle number concentrations has demonstrated that a binary homogeneous sulfuric acid-water nucleation mechanism improves simulated particle number in the FT whereas an empirical new particle formation mechanism based on sulfuric acid improves simulated particle number in the continental BL. Both the kinetic- and activation-type mechanisms equally matched the observations so our analysis was not able to determine which mechanism appears to be dominant in the atmosphere. In future work we will attempt to gain further insight into the particle formation mechanism through a detailed analysis of particle formation events including evaluation of simulated particle formation and growth rates. Because the rate of new particle formation is sensitive both to the concentration of gaseous sulfuric acid and to the pre-existing aerosol surface area [Spracklen et al., 2006] there is a need to evaluate the model at a range of sites where observations of

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both are available. A detailed analysis of particle concentrations in the free and upper troposphere is also needed to better understand particle formation in this important region of the atmosphere. Recent laboratory experiments have suggested that in addition to sulfuric acid, organics may play a role in the nucleation process under atmospheric conditions [Metzger et al., 2010]. Targeted field campaigns in a range of contrasting environments are now needed along with model studies to evaluate the contribution of this mechanism to particle formation in the real atmosphere.

Primary aerosol sources can suppress nucleation through providing a condensation sink for sulfuric acid vapor and scavenging newly formed particles through increasing the coagulation sink. Previous studies have shown that dust particles can reduce CN concentrations within dusty regions by up to 20% through this mechanism [Manktelow et al., 2010]. Future studies must therefore study the contribution of dust, primary biological aerosol particles [Heald and Spracklen, 2009] and trace metal emissions [Birmili et al., 2006] to particle surface area and the corresponding suppression of new particle formation.

The production of CCN from particle formation also depends on particle growth rates which depend greatly on the availability of SOA precursors. A better understanding of particle growth rates is therefore important to understand the role of particle formation in the production of climate relevant particles. In Merikanto et al. (2009) we use the model evaluated here to quantify the contribution of both particle formation and primary particles to regional and global CN and CCN concentrations.”

References

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