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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Aerosol indirect forcing in a global model with particle nucleation" *by* M. Wang and J. E. Penner

**M. Wang and J. E. Penner**

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**We are grateful for the evaluation of the reviewer, which have allowed us to improve and clarify the manuscript. Below we address each of the comments. The reviewer comments are in italics and our response is in bold**.

**Anonymous Referee #1**

*Wang and Penner present a GCM-based estimate of the contribution of atmospheric nucleation and primary sulphate particles on the CCN concentration and on the 1st indirect aerosol radiative effect.*

*The manuscript is well written and highlights some current and important issues related to CCN sources, and the methodology used is scientifically sound. Some of the results presented are novel and should be interesting to a range of atmospheric scientists.*

*I therefore recommend the manuscript to be published after the following comments have been addressed.*

*Specific comments:*

*1) The same authors conclude in the abstract of Wang et al. (2008, submitted to JGR) that a 3-mode representation of sulphate in their model improves the simulation results against observations and that this indicates the importance to represent the freshly nucleated particles separately. Why was a 2-mode version then chosen for this study that specifically focuses on nucleation and the growth of these particles to CCN sizes? The authors should include a discussion of how the 2-mode only version might affect their results and conclusions.*

**We thank the reviewer for bringing this question to our attention. This question was also raised by one of the reviewers for our JGR manuscript. An additional 3-mode simulation that includes boundary layer nucleation was added in Wang et al. (2008). Our results showed that the 3-mode representation has small effect on the simulated accumulation mode aerosol particles in the boundary layer. Here we added the following discussion in section 7 and also refer readers to Wang08 for more details:** "In this study, a 2-mode representation (nucleation/Aitken mode and accumulation mode) of the sulfate aerosol size distribution is used to simulate the effects of nucleation events from both boundary layer nucleation and BHN. As shown in Wang08, a 3-mode representation with an additional mode representing nucleation sizes (radius < 5 nm) produces fewer Aitken mode particles and more accumulation mode particles in the upper troposphere because of the large amount of freshly nucleated particles from BHN. However, the 3-mode representation has a smaller effect on the accumulation mode particles in the boundary layer even when boundary layer nucleation is included. This is partly due to the fact that the aerosol particles generated from boundary layer nucleation are grown by condensation up to a size of 3 nm and have lower number concentrations in the nucleation mode compared with those from BHN. In addition, primary particles are important sources for accumulation mode par-



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ticles in the boundary layer, whereas the growth of particles from the nucleation mode is important in the free troposphere. As a result, the effects of different size distribution treatments (3-mode vs. 2-mode) is small. Since accumulation mode particles in the boundary layer are the focus of this study, we would expect the 3-mode representation to have small effects on the results reported here."

*2) As also pointed out by J. Kazil in his comment, this study assumes BL nucleation to occur via activation of sulphuric acid containing clusters and uses globally the prefactor reported for one boreal forest site. However, Riipinen et al. (2007) have shown that this prefactor can differ at least by one to two orders of magnitude between different sites - and even between two campaigns for the same site! Furthermore, Spracklen et al. (2008) showed that changing this prefactor by two orders of magnitude has significant effects on the predicted CCN from nucleation. The authors do mention the uncertainty in the nucleation mechanism at the very end of the manuscript; however, I would like to see much more discussion on this topic.*

**The sensitivity of the CCN enhancement from boundary layer nucleation to different prefactors has been studied in Spracklen et al. (2008), and is not the focus of this study. Here we focus on how these effects depend on simulated primary particles. But we do bring up this point by citing Spracklen et al. (2008) in section 4. Here is the text:** "Spracklen et al. (2008) showed that the enhancement in April is 9% when the rate coefficient is 2.0x10$^{-6}$/s. Their result is close to our annual average result (5.3% enhancement in the case when primary-emitted sulfate particles are included), although we have somewhat different emissions and concentrations of other primary particles (e.g., carbonaceous aerosol, sea salt) and have a different magnitude of emissions of the precursor species ($SO_2$ and DMS) of sulfuric acid gas. They also showed that the enhancement in April ranges from 3 to 20% when the rate coefficient ranges from 2.0x10$^{-8}$ to 2.0x10$^{-4}$/s.

Our results suggest that the effects of including boundary layer nucleation on the CCN concentrations depend on both the rate coefficients (as shown by Spracklen et al.,



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2008), and in an important way on the assumed and simulated primary particles."

**We also added more discussion of this point in section 2, and also cite the study of Kuang et al. (2008) who also show that the prefactor varies with location and environmental conditions. Given our poor understanding of nucleation mechanisms, we think this empirical parameterization is a reasonable choice for representing boundary layer nucleation in global models at the present time. Here is the text in section 2:** "In Wang08, we implemented a parameterization to simulate boundary layer nucleation in the IMPACT model. This parameterization was based on an empirical fit to newly formed particles and their dependence on sulfuric acid vapor from long-term observations of aerosol formation events at Hyytiälä, Finland (Kulmala et al., 2006; Sihto et al., 2006; Riipinen et al., 2007). This parameterization also fits nucleation events measured in a variety of continental and marine atmospheric environments (Kuang et al., 2008). The nucleation rate of 1 nm particles is first calculated using the parameterizations from Kulmala et al. (2006) and Sihto et al. (2006) as:

$j_{1nm} = A \mathrm{x} [H_2SO_4]$, (1)

or $j_{1nm} = K \mathrm{x} [H_2SO_4]^2$, (2)

where A and K are rate coefficients. In the model, 3 nm particles are added to the nucleation mode at each time step (which is dynamically determined based on the accuracy of the solution) instead of 1 nm particles. The rate of formation of 3nm particles (j3nm) is calculated from j1nm using the formula from Kerminen and Kulmala (2002).

For the rate coefficients A and K, we adopt the median values derived from the case studies in Sihto et al. (2006) which were 1.0x10$^{-6}$/s and 1.0x10$^{-12}$cm$^3$/s, respectively. Wang08 showed that, the inclusion of these boundary layer nucleation mechanisms improved the comparison of simulated aerosol size distributions with observations in the MBL. Furthermore, the use of Eq. (1) and Eq. (2) gave similar results, especially in terms of the number concentration of the accumulation mode particles. Here, we choose Eq. (1) to represent boundary layer nucleation. The contribution of BHN and



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the nucleation mechanism represented by Eq. (1) to the CCN concentration and to the first aerosol indirect forcing will be quantified.

Although the form of the empirical parameterization for nucleation is generally applicable, variations of several orders of magnitude in the prefactors A and K have been observed, depending on location and the environmental conditions (Sihto et al.; 2006; Riipinen et al., 2007; Kuang et al., 2008). This indicates that species other than sulfuric acid may play a role in nucleation. For example, biogenic iodine oxides (O'Dowd et al., 2002b), organic species (Zhang et al., 2004), and ammonia (Korhonen et al., 1999) can be important in particle formation. In addition, ion mediated nucleation may be also important (Yu et al., 2008a). Nevertheless, this simple empirical parameterization is still useful for exploring the effects of boundary layer nucleation in global models, given our poor understanding of the mechanisms behind particle formation."

*Technical comments:*

*- p. 13945, line 8 and 9: change , condensation to , condensation and wet removal, and e.g. to i.e.*

**DONE.**

*- p. 13945, line 20: Laaksonen et al. (2005) measurements are from Po Valley, a highly industrialized and polluted region in Italy - not from a sub-Arctic boreal forest.*

**We moved this reference to continental sites. Now the text reads:** "New particle formation events capable of producing CCN-size particles have been observed at locations including the sub-Arctic boreal forest (Lihavainen et al., 2003), coastal areas (O'Dowd et al., 2002a), in continental (McNaughton et al., 2004; Laaksonen et al., 2005) and cloud outflow regions (Twohy et al., 2002), and in the upper free troposphere (Singh et al., 2002)."

*- I suggest the use of term global aerosol models to refer simultaneously to both GCM and CTM based models. The currently used term global transport models may give*



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*the impression that models discussed are all CTM based. (E.g. top of page 13947)*

**DONE.**

*- p. 13948, lines 2 and 7: change Shito to Sihto and Hyyti?li? to Hyyti?l?*

**DONE.**

*- p. 13948, line 18 (and again on p. 13960, lines 12-19 ): Spracklen et al. (2008) does include primary sulphate although this is not explicitly mentioned in their paper (personal communication with Dom Spracklen).*

**We thank the reviewer for this information. We checked with Dr. Spracklen, and found that Spracklen et al. (2008) does include primary sulfate which is emitted with an aged size distribution, although this is not described in Spracklen et al. (2008). We added this information to our text:** "Spracklen et al. (2008) used the empirical nucleation rate represented by Eq. (1) but with different rate coefficients to study the effects of the boundary layer nucleation on CCN concentrations in spring (March-May) and how these depend on rate coefficient. In their model, 2.5% of the anthropogenic SO2 emissions were emitted as primary sulfate with the aged size distribution that was suggested for the AEROCOM emissions inventory (personal communication, D. Spracklen; Dentener et al., 2006). The majority (88%) of the primary anthropogenic SO2 comes from the industrial sector and is emitted at a diameter of 1 $\mu m$, while the remaining fraction comes from the traffic sector and is emitted at a diameter of 30 nm. "

*- p. 13954, line 5 and Figure 1: change nucleation parameterization to cloud activation parameterization or to cloud nucleation parameterization.*

**Done. It is changed to** "cloud activation parameterization";

*- p. 13955, line 6: Are both equation numbers wrong? If not, explain in more detail as I cannot follow the logic here.*



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**Yes, they are wrong. Both of them are Eq. (3). Changed.**

*- p. 13958, line 3: Use consistently either Wang08 or Wang et al. (2008) throughout the manuscript.*

**Wang08 is now used.**

*- p. 13962, line 27: change decrease of 6% to decrease of 6 percentage points (also p. 13963, line 14; p. 13965, lines 22 and 23).*

**Done.**

*- p. 13966, line 14 onwards: It is quite optimistic to say that BHN_PAR agrees well with the satellite data. In addition to 20-60 degrees S, there are large discrepancies over desert areas, over northern Eurasia etc.*

**We changed our statement there, and it is now:** "The magnitude and spatial distribution of cloud-top droplet number from the BHN_PRIM case agrees better with the MODIS data than that from the BHN case."

**(Note: the case names are changed following the suggestion of Referee #2).**

*- p. 13966, lines 26-27: In my opinion even a more important point is whether including BL nucleation improves the comparison compared to BHN_PAR case.*

**Yes, we agree. We put "BHN" there by mistake. It is changed to "BHN_PRIM"**

*- p. 13970, line 5: change Lohmman to Lohmann*

**Done.**

*- p. 13973, line 1: what does smaller increase and larger decrease refer to?*

**We clarified this point and now it reads:** "This suggests that including a boundary layer nucleation scheme would have lead to a smaller increase and a larger decrease in the anthropogenic fraction of CCN than that in this study, if this ion-mediated nucleation mechanism were used and the increase in temperature from the PI to the PD were



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included."

*- Table 1: give units for Ni. Give Ni for the largest dust mode as 7.3e-5 to be consistent.*

**7.3e-5 is used. A footnote is added for Ni:** "[a]Ni is the fraction of the total particle number in a given size range and is dimensionless."

*- Table 2: what do the hygroscopicity values refer to? Please explain in more detail.*

**This is now explained in the following footnote in Table 2:** "[a]The hygroscopicity parameter depends on the number of dissolved ions per molecule, the osmotic coefficient, the soluble mass fraction, the component density, and the molecular weight, as defined by Eq. (3) in Abdul-Razzak and Ghan (2000), and values are taken from Ghan et al. (2001b)."

*- Table 3: superscript f should be one line lower (BHN_PAR, not BLBHN). I also agree with Referee #2 that the simulation names are hard to follow.*

**This is now changed. Following the suggestions of Referee #2, we changed case names (BL1st to EMP, and PAR to PRIM).**

*- Figures 2 and 6: Y axis label is incomprehensible*

**We changed Y axis label to "Approximate Pressure (hPa)", and we added the following text in the caption of Figure 2:** "CAM3 used a hybrid vertical coordinate and the pressure at the kth model level is given by $p(k) = A(k)p_0 + B(k)p_s$, where $p_s$ is surface pressure, $p_0$ is a specified constant pressure (1000 hPa), $A$ and $B$ are coefficients. Data are plotted as a function of the hybrid vertical coordinate times 1000 and labeled Approximate Pressure."

**We also found that Figure 6 was not plotted correctly and that only the results in December was plotted previously. Here we corrected Figure 6, and updated texts in Section 5 correspondingly (see paragraph 2 and 6 in section 5).**

*- Figure 3 and 4 captions, and in text: refering to this model level as 930 hPa is much*



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*more informative than calling it the third model level*

**This is done. 930 hPa is used, and we put "the third model level" in the parenthesis.**

*- Figure 9: explain in caption that the values given in the title are global averages. Also in caption write reference to panels (a) and (b) before the explanations of figure content (the same comment applies to Fig. 11)*

**Done.**

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Interactive comment on Atmos. Chem. Phys. Discuss., 8, 13943, 2008.