**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "

Air pollution control and decreasing new particle formation lead to strong climate warming" *by* R. Makkonen et al.

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This paper uses the modal microphysics algorithm, M7, coupled to ECHAM5-HAM to explore how aerosol radiative forcing might evolve in the future according to emissions forecast in the new RCPs. A key result, highlighted in the title and the abstract, is that nucleation contributes significantly to the indirect effect in these results, and that this contribution will decrease with future emissions changes. Therefore, a proper accounting of nucleation, might make the climate warming associated with aerosol

controls worse than previously estimated.

Overall, the results are quite interesting but are seriously lacking in discussion and context. I recommend major revisions to document more completely the model used, the results attained, and to compare your results to similar work in the literature. I will note that my assessment of the "scientific quality" as "poor" is not because I think the work is poor science per se. ACP includes "are the results discussed in an appropriate and balanced way (consideration of related work)" here, and it is the documentation and discussion I find lacking.

1) Other literature: First, the question of how much nucleation does or does not contribute to CCN, CDNC, and the indirect effect has seen a number of publications lately. The results shown here seem to be on the high end of other estimates although not necessarily inconsistent. This can be confusing for non-modelers, who simply say "this paper shows a 10% effect but this one has a factor of 2 or 3". The published results are probably more consistent than would appear at first glance because the numbers quoted are for different sensitivity cases. For example, it appears that the effects are much larger when comparing some nucleation to no nucleation at all than when comparing two different nucleation schemes. It also seems to make a difference whether nucleation is perturbed over the entire troposphere or boundary layer only. A partial list of noteworthy papers in this regard are: [Merikanto et al., 2010; Merikanto et al., 2009; Pierce and Adams, 2009; Spracklen et al., 2006; Spracklen et al., 2008; Yu et al., 2008]. Although most or all of these are cited somewhere in the paper, there is no real effort made to compare these results to the rest of the literature. This is a shame since it propagates the confusion. I strongly urge you to add a paragraph to the discussion section with a quantitative discussion comparing where your results are larger or smaller than other estimates and suggest reasons for differences.

1a) On a related note, some of the differences between published results likely depend on the assumptions made about primary aerosol emissions. The model description section (which is extremely short) needs to document/review these, and point the

reader to the correct earlier work for details.

1b) Similarly, it would be extremely useful to see the CN and CCN concentrations produced by the model with nucleation turned off. This would also help the reader assess the strength of primary emissions used in the model. These could be easily added to Figure 1 (or to a separate analogous figure).

2) Document and critique activaction scheme: Although critical to the results, the scheme for activating particles into CCN is not fully documented and appears to favor particles that are really too small. Little is said about the activation scheme in the current paper, but it appears to be the same one used and discussed in Makkonen et al. [2009] and first developed in Lohmann et al. [2007]. In fairness to the current authors, most of the following questions probably should have been already addressed in the Lohmann paper. According to those references, the activation of cloud droplets depends on the number of particles with wet radius larger than 35 nm. There are several points here that are not clear or troublesome.

2a) This is wet radius at what RH value? The model ambient value? 80? It is unclear but should have an important effect on the number of activated particles.

2b) A wet radius of 35 nm is on the small side for what can be considered CCN under atmospheric conditions. Converting to diameter and assuming a diameter growth factor of 1.4 (e.g. [Gasparini et al., 2006]), this corresponds to a dry diameter of 50 nm. This is small for stratiform clouds that should contribute most to the aerosol indirect effect. Even for a very high kappa of 1, it takes a supersaturation of excess of 0.3% to activate a 50 nm particle (e.g. see Figure 1 of [Petters and Kreidenweis, 2007]). Using my favorite numbers, supersaturation of 0.2% for stratiform clouds and a kappa of 0.5 (a mixture of sulfate and organics), a particle should have a dry diameter of about 90 nm to activate. Since the effect of nucleation on the aerosol size distribution will be most pronounced in the smaller size range, the use of a small activation diameter will tend to enhance (artificially) the effect of nucleation on CCN. How would the results differ if

C12725

a larger (probably more realistic) activation diameter were used?

2c) Second, since this cutoff size falls in the middle of the Aitken mode, how is the size interpolation done to determine the number of particles from the Aitken mode that activate? Do all the Aitken mode particles activate (or not) together depending on the mode's average size? Is there a lognormal distribution fit to the mode and then just the fraction that exceeds 35 nm wet radius contributes to CCN? I do not see this, not even in the original Lohmann et al. [2007], but see Korhonen et al. [2005] for the importance of doing some kind of interpolation in this regard in sectional models (the same should apply to modal models).

2d) "aerosols are activated as cloud droplets with a cut-off radius of 35 nm." This description of the activation parameterization is too short given its importance to the paper and also imprecise. In the Lohmann 2007 paper, not all particles larger than 35 nm wet radius activate per se. The number of such particles is the input to the parameterization, and then there is an empirical function that says what fraction actually activate, implicitly allowing particles to compete for water vapor. Please provide a more detailed and accurate summary of how particles are activated in the model (e.g. a short paragraph).

2e) Why use an empirical activation scheme that treats all particles with wet radius larger than 35 nm as equal? Larger particles (i.e. primary particles) can compete more effectively for water vapor, but the scheme used here treats any particle larger than that threshold as the same. Why not use a more physically realistic parameterization? These are readily available.

3) Size resolution of the modal approach: Several papers now discuss the microphysics that govern the "survival probability" for fresh nuclei to grow to CCN sizes. Essentially, a particle must grow by condensation before it undergoes coagulational scavenging by a larger particle [Kerminen et al., 2004a; Kerminen et al., 2004b; Kuang et al., 2009; Pierce and Adams, 2007]. Therefore, a lot depends on the model's ability to represent

C12726

accurately the coagulation of smaller particles before they become CCN and maintain the distinction between CCN and non-CCN particles. Although the results shown here do not seem to be completely outside the range of other results, they do appear to be on the high end of the range. There are a few places where the modal approach may cause a high bias that should be documented/reviewed to put these results in context.

3a) Please briefly review the size configuration of the modes used in M7, since the details are relevant to the results presented. Related to the earlier point about activation, which modes are eligible to activate? Although details are unclear, it appears that the Aitken mode does activate, although this includes particles as small as 10 nm diameter.

3b) There is potential for "numerical diffusion" within the Aitken mode, which covers 10 nm to 100 nm particle diameter. For example, adding to the Aitken mode a 10 nm particle (e.g. just grown from the nucleation mode) plus a primary particle of 80 nm is equivalent to adding two 63 nm particles since they give the same number and mass. In other words, recently nucleated particles moving into the Aitken mode are free to "borrow" mass from larger particles in the same mode. I don't see an easy way to remedy this or quantify the potential bias, but I do think it should be noted.

3c) The modal approach used here is a "pseudomodal" approach in which "the rate constants for coagulation and condensation are calculated for the average mode radius rather than the integral over the mode." (see paragraph 10 of Vignati et al. [2004] as well as eqn 16 of Stier et al. [2005]). Whereas the coagulation coefficients of real particles within the Aitken mode varies by approximately an order of magnitude, this is not rigorously accounted for. This means that, as soon as a particle with $D_p = 10$ nm moves into the model's Aitken mode, it is treated like any other Aitken particle for purposes of coagulation. If the average particle size in the Aitken mode is 30 to 50 nm, then its rate of coagulational scavenging is under-estimated by approximately a factor of 3 to 5. A similar argument applies to a freshly nucleated particle entering the nucleation mode. The tendency to under-predict the coagulation of the smaller

particles should artificially increase their survival probability in the model. It is true that this might be partly compensated by the opposite tendency for particles larger than the average mode size. At least, since a 50 nm dry diameter particle can be a CCN, the tendency to help them survive the 10 to 50 nm range already enhances the effect of nucleation of CCN.

4) The abstract is very short (probably the shortest I have ever seen) and should be used more effectively. For example, although the contribution of nucleation to the forcing is a key result of the paper, the forcing values with and without nucleation are not even quantified in the abstract.

5) "We use the global climate model ECHAM5-HAM (Stier et al., 2005) with a novel description of boundary layer nucleation and particle growth by BVOC oxidation products (Makkonen et al., 2009)." What is the novel description of boundary layer nucleation? Later, it says that the manuscript uses the "activation-type" nucleation of Kulmala et al. 2006. Either the treatments of nucleation and SOA are not so novel (they are already used in Makkonen et al 2009, not to mention several other modeling studies) or any novelty needs to be documented here.

6) The forcing values need better documentation in several ways:

6a) With regards to "total" aerosol forcing, what does total mean? Direct and indirect? Is the "second" indirect effect included? The semi-direct effect?

6b) Similarly, the discussion of forcings in the RCP scenarios needs to be more precise. There are several mentions of "total" forcing, but I think these mean long-lived GHGs. I.e. ozone and aerosols are not included.

6c) The abstract claims that "The total aerosol forcing ($-1.61 Wm-2$ in year 2000) is simulated to be greatly reduced in the future, to $-0.23 Wm-2$, mainly due to decrease in SO2 emissions and resulting decrease in new particle formation." However, the attribution of forcing to aerosol species or to the indirect effect is non-existent in the

paper, so this statement is largely unsubstantiated. The reader's assessment of the results would benefit greatly from a table that breaks the forcing down by type. As far as I can tell, every aerosol forcing includes at least direct and indirect effects together. Since a lot of the key results revolve around nucleation, CCN, and the indirect effect, at the very least, there should be some quantification of the indirect effects alone.

6d) Don't changes in POA emissions play any role in the changes in forcing? I would imagine that there are changes in the direct radiative forcing from OA that are playing a significant role here, but I have no means to assess how much of a role they are playing.

7) p. 25996, lines 6-9: "At global scale, only about 5% of the future decrease in CN concentration can be explained by changes in primary particle emissions, the rest being due to a decrease in anthropogenic SO2 emissions and the resulting decrease in atmospheric new particle formation." I see nothing in the paper that describes how is this "explanation" done. Since aerosol number concentrations are a nonlinear function of nucleation and emissions due to coagulation, condensational growth etc, attributing number to nucleation or primary particles is non-trivial.

8) p. 25996, "Our model shows an increase of 20% in CDNC from year 1750 to 2000, which is lower than previously reported estimates" and then, "This suggests that our model is less sensitive to the anthropogenic influence since pre-industrial times." It is perplexing how this is consistent with the forcing values and sensitivity to nucleation. If the CDNC change is small, why is the total aerosol forcing (year 2000 versus pre-industrial) equal to -1.6 W/m2, i.e. a pretty strong aerosol forcing compared to IPCC ranges? Is there more direct effect than indirect effect (again, a breakdown would help the reader understand what's going on)? If CDNC changes (and presumably indirect effects) are modest, how does the nucleation-CCN link end up being so strong? Is the model somehow getting a much stronger indirect effect from a more modest CDNC change compared to other models?

C12729

9) What do the error bars in Figure 3 represent?

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C12731