Atmos. Chem. Phys. Discuss., 11, C14263–C14276, 2012
www.atmos-chem-phys-discuss.net/11/C14263/2012/

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.

R. Makkonen et al.

risto.makkonen@helsinki.fi

Received and published: 15 January 2012

"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."

We agree with Prof. Adams that several ascpets in the manuscript could be extended. In the revised version, we have taken into account the specific issues raised below.

"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 10are 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."

Several papers in literature compare simulations with and without boundary nucleation, but usually lack the comparison against the case of no nucleation. We have improved the comparison to existing literature in the revised manuscript, taking better into account the used nucleation mechanisms and analyzed variables (CCN, CDNC). We think that our results are rather sensitive to nucleation, but in good agreement with literature.

"1a) On a related note, some of the differences between published results likely depend on the assumptions made about primary aerosol emissions. The model descrip-

ACPD
11, C14263–C14276, 2012

tion section (which is extremely short) needs to document/review these, and point the reader to the correct earlier work for details."

We have rewritten the model description, and included a description on the primary emission size distribution.

"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). "

We have extended the figures to include CN and CCN from experiments without nucleation. We have also added a table to present global averages of CN, CCN and CDNC.

"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. "

The aerosol-cloud coupling is described in detail in Lohmann et al. (2007) but deserves more documentation in this manuscript. We have extended the method description to include the most important features of cloud droplet activation scheme.

"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. "

The wet radius refers is calculated online in the model at ambient RH. For particles con-

ACPD

11, C14263–C14276, 2012

taining sulfate, water vapour equilibrium is based on a generalised form of the Kelvin equation. For sea-salt containing particles, the efflorescence relative humidity is taken into account. More details can be found from Stier et al. (2005). We have improved the description in the manuscript.

"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 0.3a 50 nm particle (e.g. see Figure 1 of [Petters and Kreidenweis, 2007]). Using my favorite numbers, supersaturation of 0.2mixture 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 a larger (probably more realistic) activation diameter were used?"

The droplet activation radius is in the middle of the Aitken mode, and hence the droplet activation is very sensitive to number concentration changes in Aitken mode. The Aitken mode number concentration is sensitive to both new particles growing from nucleation mode and primary emissions. In this study we implemented primary emission sizes according to Stier et al. (2005), where the primary emission radii are doubled compared to the AeroCom radii (Dentener et al. 2006). This reduces the sensitivity of cloud droplet activation to primary emissions. If the droplet activation is "too" sensitive to nucleated particles, the sensitivity naturally affects clouds also in pre-industrial time.

The activation radius of 35 nm is between observed continental and maritime radius (Dusek et al., 2006).

Although we are fully aware of the limitations of using an empirical formulation of cloud

ACPD
11, C14263–C14276, 2012

droplet activation in a global model, the LinLeaitch scheme is shown to perform well in ECHAM5-HAM, producing for example the cloud albedo effect similar to average IPCC value (Forster et al., 2007).

"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). "

The aerosol modes are assumed to be lognormal, and error function is used to determine the number of particles larger than 35 nm, hence the whole Aitken mode is not activated.

"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). "

The cloud droplet activation is an important part of the manuscript, and hence has been decribed more clearly in the revised manuscript.

"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

ACPD
11, C14263–C14276, 2012

that threshold as the same. Why not use a more physically realistic parameterization? These are readily available."

Mechanistical approaches for cloud droplet activation are available, e.g. Abdul-RazzakGhan (2000) and FountoukisNenes (2005). However, the activation scheme LinLeaitch was the only one implemented and evaluated in ECHAM5-HAM.

We have recently tested the sensitivity of our results to the cloud activation scheme by additional simulations with Abdul-RazzakGhan (2000) scheme. We have found that the main results of the manuscript (changes in anthropogenic forcing) are very similar with the two activation schemes. To our knowledge, there exists no thorough global model comparisons of different activation schemes in the literature, which makes it difficult to analyze the uncertainties of this study.

"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 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."

Including aerosols in a global climate model is a compromise between computational efficiency and the ability of the model to reproduce the essential microphysics. The modal model setup used in this study is widely used global models: in an AeroCom intercomparison (Textor et al., 2006) half of the models were based on a sectional method and half on modal method. We agree the limitations of a modal approach, and

ACPD

11, C14263–C14276, 2012

have included some specific points in the manuscript.

"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."

As explained earlier, the model assumes that only a part of the Aitken mode (wet radii>35 nm) is able to activate as cloud droplets. We have included a description of the modal setup in the manuscript.

"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."

We are aware that this is one of the drawbacks in the modal setup used. We have included some discussion of this problem.

"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 Dp = 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

ACPD
11, C14263–C14276, 2012

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."

This is one of the problems with modal approaches which is likely to affect our results to some extent, but we are unable to access the importance in the framework of the current study.

"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."

We have expanded the abstract to cover the main results of the manuscript.

"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."

The word novel is removed from the model description.

"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?"

ACPD
11, C14263–C14276, 2012

The total aerosol forcing reported in the manuscript is calculated as a radiative flux perturbation (Haywood et al., 2009; Lohmann et al., 2010) at the top-of-atmosphere. The flux perturbation is calculated for total radiation (SW+LW). The forcing includes direct, semi-direct, 1st indirect effect and 2nd indirect effect. While the climate is kept fixed with prescribed montly sea-surface temperatures (same for all simulations), the clouds can adjust to changes in cloud droplet number via changes in precipitation, cloud lifetime and liquid water path. Hence, the second indirect effect is included in the calculations. We have extended the description of forcing calculations in the manuscript.

"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."

We have modified the description of the RCP scenarios. In this paper, the reported total aerosol forcing includes the direct and both first and second indirect effects, but excludes forcing from other sources (LLGHGs, etc.). In the RCP description the total forcing included also LLGHGs.

"6c) The abstract claims that “The total aerosol forcing (-1.61Wm2 in year 2000) is simulated to be greatly reduced in the future, to -0.23Wm2, 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."

The model setup is used to obtain total aerosol forcing, and the separation of different effects is not straightforward. To give some idea on the magnitude of the direct effect, we have included the clear-sky direct aerosol radiative forcing in the revised

ACPD

11, C14263–C14276, 2012

manuscript. However, even the clear-sky direct effect is affected by changes in cloud microphysics and it is not strictly a forcing. We do not expect a significant modification to direct effect by nucleated particles themself, but the change in aerosol size distribution can induce secondary effects that influence the aerosol optical depth.

"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."

The applied emission inventories include changes in POA. The effect of POA on both direct and indirect effects is included in the model, but the strength is not quantified separately per compound in this study. The direct radiative forcings of RCP projections are studied in e.g. Lamarque et al. (2011), where the organic carbon present-day clear-sky radiative forcing of -0.05 W m^{-2} is reduced until 2100, ranging from -0.04 to 0 W m^{-2}.

"7) p. 25996, lines 6-9: "At global scale, only about 5concentration 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."

The claim is rephrased in the revised manuscript to provide a comparison between global CN numbers with and without nucleation. The fraction of 5

"8) p. 25996, "Our model shows an increase of 20which 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

ACPD
11, C14263–C14276, 2012

is the total aerosol forcing (year 2000 versus preindustrial) 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?"

The low sensitivity referred only to the CDNC, not the forcing. The CCN is more sensitive than CDNC to anthropogenic influence, and the comparison to previous results includes both CCN and CDNC. Also, the relative increase of CCN/CDNC from preindustrial to present-day is highly sensitive to the absolute values. We have revised the comparison to earlier results. The total aerosol forcing reported here includes the direct and semi-direct effects, and both first and second indirect effects. The present-day total aerosol forcing (-1.6 W m^{-2}) is in good agreement with previous model studies: average total SW aerosol forcing of -1.53$\pm$0.60 W m^{-2} from 10 different GCMs (Quaas et al., 2009), aerosol climate forcing of -1.6$\pm$0.3 W m^{-2} inferred from Earth's energy balance (Hansen et al., 2011). Many models only include 1st indirect effect. It is likely that the 2nd indirect effect included in our study makes the present-day aerosol forcing more sensitive to the increase in CDNC, and we are looking into this in ongoing research.

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

Error bars in Fig. 3 were one standard deviation for present-day and a combination of standard deviation and emission scenario variation for year 2100. We have redrawn the figure to ease the interpretation.

References used in the author's response:

Abdul-Razzak, H. and Ghan, S. (2000). A parameterization of aerosol activation, 2, Multiple aerosol types. Journal of Geophysical Research 105(D5): doi:

ACPD

11, C14263–C14276, 2012

10.1029/1999JD901161. issn: 0148-0227.

Dentener, F., Kinne, S., Bond, T., Boucher, O., Cofala, J., Generoso, S., Ginoux, P., Gong, S., Hoelzemann, J. J., Ito, A., Marelli, L., Penner, J. E., Putaud, J.-P., Textor, C., Schulz, M., van der Werf, G. R., and Wilson, J.: Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed data-sets for AeroCom, Atmos. Chem. Phys., 6, 4321-4344, doi:10.5194/acp-6-4321-2006, 2006.

Dusek, U., Frank, G. P., Hildebrandt, L., Curtius, J., Schneider, J., Walter, S., Chand, D., Drewnick, F., Hings, S., Jung, D., Borrmann, S., and Andreae, M. O.: Size matters more than chemistry for cloud-nucleating ability of aerosol particles, Science, 312, 1375–1378, 2006.

Fountoukis, C. and Nenes, A. (2005). Continued development of a cloud droplet formation parameterization for global climate models. Journal of Geophysical Research 110(D11): doi: 10.1029/2004JD005591. issn: 0148-0227.

Forster, P., Ramaswamy, V., Artaxo, P., Berntsen, T., Betts, R., Fahey, D., Haywood, J., Lean, J., Lowe, D., Myhre, G., Nganga, J., Prinn, R., Raga, G., Schulz, M., and Van Dorland, R.: Radiative Forcing of Climate Change, in: Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K. B., Tignor, M., and Miller, H., Cambridge Univ. Press, New York, pp. 129–234, 2007.

Hansen, J., Sato, M., Kharecha, P., and von Schuckmann, K.: Earth's energy imbalance and implications, Atmos. Chem. Phys., 11, 13421-13449, doi:10.5194/acp-11-13421-2011, 2011.

Haywood, J. M., Donner, L. J., Jones, A., and Golaz, J.-C.: Global indirect radiative forcing caused by aerosols: IPCC (2007) and beyond, in Clouds in the Perturbed Climate System, edited by J. Heintzenberg and R. J. Charlson, pp. 451–467, MIT Press,

ACPD

11, C14263–C14276, 2012

Cambridge, 2009

Lamarque, J.-F., Kyle, G., Meinshausen, M., Riahi, K., Smith, S., van Vuuren, D., Conley, A., and Vitt, F.: Global and regional evolution of short-lived radiatively-active gases and aerosols in the representative concentration pathways. Climatic Change, 109:191–212, 10.1007/s10584-011-0155-0, 2011.

Lohmann, U., Rotstayn, L., Storelvmo, T., Jones, A., Menon, S., Quaas, J., Ekman, A. M. L., Koch, D., and Ruedy, R.: Total aerosol effect: radiative forcing or radiative flux perturbation?, Atmos. Chem. Phys., 10, 3235-3246, doi:10.5194/acp-10-3235-2010, 2010.

Lohmann, U., Stier, P., Hoose, C., Ferrachat, S., Kloster, S., Roeckner, E., and Zhang, J.: Cloud microphysics and aerosol indirect effects in the global climate model ECHAM5-HAM, Atmos. Chem. Phys., 7, 3425-3446, doi:10.5194/acp-7-3425-2007, 2007.

Quaas, J., Ming, Y., Menon, S., Takemura, T., Wang, M., Penner, J. E., Gettelman, A., Lohmann, U., Bellouin, N., Boucher, O., Sayer, A. M., Thomas, G. E., McComiskey, A., Feingold, G., Hoose, C., Kristjánsson, J. E., Liu, X., Balkanski, Y., Donner, L. J., Ginoux, P. A., Stier, P., Grandey, B., Feichter, J., Sednev, I., Bauer, S. E., Koch, D., Grainger, R. G., Kirkevåg, A., Iversen, T., Seland, Ø., Easter, R., Ghan, S. J., Rasch, P. J., Morrison, H., Lamarque, J.-F., Iacono, M. J., Kinne, S., and Schulz, M.: Aerosol indirect effects – general circulation model intercomparison and evaluation with satellite data, Atmos. Chem. Phys., 9, 8697-8717, doi:10.5194/acp-9-8697-2009, 2009.

Stier, P., Feichter, J., Kinne, S., Kloster, S., Vignati, E., Wilson, J., Ganzeveld, L., Tegen, I., Werner, M., Balkanski, Y., Schulz, M., Boucher, O., Minikin, A., and Petzold, A.: The aerosol-climate model ECHAM5-HAM, Atmos. Chem. Phys., 5, 1125-1156, doi:10.5194/acp-5-1125-2005, 2005.

Textor, C., Schulz, M., Guibert, S., Kinne, S., Balkanski, Y., Bauer, S., Berntsen, T.,

ACPD

11, C14263–C14276, 2012

Berglen, T., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R., Feichter, H., Fillmore, D., Ghan, S., Ginoux, P., Gong, S., Grini, A., Hendricks, J., Horowitz, L., Huang, P., Isaksen, I., Iversen, I., Kloster, S., Koch, D., Kirkevåg, A., Kristjansson, J. E., Krol, M., Lauer, A., Lamarque, J. F., Liu, X., Montanaro, V., Myhre, G., Penner, J., Pitari, G., Reddy, S., Seland, Ø., Stier, P., Takemura, T., and Tie, X.: Analysis and quantification of the diversities of aerosol life cycles within AeroCom, Atmos. Chem. Phys., 6, 1777-1813, doi:10.5194/acp-6-1777-2006, 2006.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 25991, 2011.