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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Particle number concentrations over Europe in 2030: the role of emissions and new particle formation" *by* L. Ahlm et al.

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Response to comments by reviewer 3

We thank reviewer 3 for comments and suggestions for improvement of our manuscript. The comments from the reviewer followed by our responses to the comments can be seen below.

Comment 1: The nucleation discussion in the introduction leaves the reader with the idea that ternary H2SO4-NH3-H2O nucleation is the perfect choice for modeling nucleation in a transport model. Stating that a scaled nucleation rate parameterization can

predict measured number concentrations seems rather vague. It is generally accepted that we are missing many details on the first steps of atmospheric new particle formation, but several parameterizations are available for modeling purposes. The most used one, at least in global aerosol models, is perhaps activation-type nucleation (Kulmala et al., 2006). The role of ions should also be briefly mentioned in the paragraph. Organic vapours can also affect nucleation rates (e.g. Paasonen et al. 2010, Riccobono et al. 2012 and references therein), which might play a role in the future evolution of number concentrations if biogenic emissions of organic vapours are changing with climate.

Response: We agree with the reviewer that the H2SO4-NH3-H2O nucleation parameterization that we use is not perfect and very likely miss components that may be important. We also stated in Sect. 2 that "While the parameterization of Napari et al. (2002) might not provide the right mechanistic picture, it may still be a reasonable approximation due to ammonia being a tracer for other bases, e.g. amines." However, we agree with the reviewer that the introduction benefits from some more details on nucleation and we have therefore added some more discussion on nucleation and parameterizations, e.g. ion-induced nucleation, activation theory, and potential roles of amines and organics. We now also mention that the reason that we use the scaled ternary H2SO4-NH3-H2O nucleation parameterization is that Fountoukis et al. (2012) compared this parameterization with activation and kinetic nucleation parameterizations and found that the scaled ternary nucleation parameterization agreed best with observations over Europe.

Comment 2: The simulated timescale of one month limits the generality of the results, which is mentioned in some locations of the manuscript. This might not be as problematic if the identical model was previously evaluated against results from a full-year simulation, but also Fountoukis et al. (2012) presents results from May only. Based on one-month evaluation, the reader cannot have a thorough view of the model performance in the European domain. Also, as mentioned, conclusions would be a lot more profound with full-year simulations.

Response: In the current stage we only have evaluated emission input files for May 2008. Unfortunately, the development and evaluation of number emission inputs files for other seasons requires a considerable amount of effort and time. For example, the development of winter number emissions requires dealing with the uncertainties of wintertime wood-burning emissions. Our work has suggested that these emissions are underestimated in several European countries and as the wood-burning source dominates wintertime number emissions the corresponding emission inventory will be very uncertain. As a result we needed to restrict our current work to the simulated period. To investigate how new particle formation depends on meteorology we performed a new set of simulations where we used emission input files from May 2008 combined with meteorology files from January 2010. These tests allowed the quantification of meteorology on the particle production assuming constant emissions. These simulations resulted in Ntot concentrations that were approximately a factor of five lower than those obtained in the original simulations. The lower particle number concentrations are the result of reduced new particle formation. We now state in Sect. 4.5 that the fact that we have focused on a photochemically active period in this study implies that the estimate of the contribution of new particle formation to the particle number concentration represents an upper limit for the influence of new particle formation. This also means that the sensitivity of the particle number concentration to SO2 emissions also probably represents an upper limit for this sensitivity.

Comment 3: If only one month is simulated, is there any spin-up period to allow for distribution of gases and particles? Is the atmosphere initiated empty with respect to aerosols and gases? It is mentioned that the first two days are excluded from the results, but this is not enough for any stabilization of upper troposphere. Merikanto et al. (2009) showed that upper tropospheric nucleation (UTN) can contribute up to 20-25% of ground-level CN (>3nm) over certain regions in Europe (the effect is even larger over oceans). If PMCAMx-UF is initiated with an empty atmosphere and non-existing spin-up period, the contribution of UTN on ground-level concentrations is most likely underestimated.

Response: PMCAMx-UF is not initiated with an empty atmosphere, but instead with reasonable concentrations of all species. We have made tests removing more than two days, i.e. a longer spin-up period, but this did not significantly change the output results. This is now mentioned in the revised paper.

Comment 4: With possibly long lifetime of aerosols, the application of constant lateral boundary conditions for aerosols is a clear limitation of model simulations. Presumably, the effect of boundary conditions on ground-level aerosol concentration in central Europe is small, but it is difficult for the reader to see where transport from boundaries could even dominate the results. The transport from outside Europe is also related to the contribution of upper tropospheric nucleation on ground-level concentrations.

Response: We agree that the applied boundary conditions naturally add some uncertainty to the model simulations, and are definitely a simplified representation of long-range transport. However, the relatively good agreement between the observed and simulated aerosol concentrations and trends at the EUCAARI sites gives confidence of the simulated processes and emissions governing aerosol number concentrations. We thus believe the model should give a reasonable insight into the sensitivities of particle number concentrations in the European boundary layer. We also agree that the contribution of upper tropospheric (UT) nucleation processes on the ground-level concentrations is one of the important questions to be resolved in the future – which requires for e.g. accurately accounting for the aerosol-cloud-aerosol interactions and vertical (and horizontal) transport patters. A detailed look at the vertical transport processes and UT nucleation processes in PMCAMx-UF is a topic that we are currently working on. We did not change the concentrations at the model boundaries for the different scenarios. We have added this information to the manuscript.

Comment 5: Why were the emissions of aerosols and trace gases only scaled by a constant factor? This seems like a drastic oversimplification as more detailed information is included in the IIASA scenarios. Could a similar scaling have been used for aerosol concentrations at model boundaries?

Response: All the scenarios in the IIASA report do not have emissions separated by country, and we wanted to be able to compare the different emission scenarios and their resulting changes in particle number concentrations. Furthermore, if one wants to investigate how sensitive the particle number concentration is to emission reductions in different areas it is better to apply a constant factor. We did not change the aerosol concentrations at the boundaries for the different scenarios. We have added this information to Sect. 3.2.

Comment 6: The manuscript includes three scenarios: baseline (current legislation), maximum technically feasible reductions and maximum control efforts. Do the authors agree that these scenarios cover the expected range of future emissions, or would there also be room for a more pessimistic scenario?

Response: The baseline scenario represents the most likely estimate by IIASA, whereas the MTFR and MCE scenarios represent emission reductions that would be possible but perhaps less likely. We think it is still important to investigate how the particle number concentration would respond to these more optimistic scenarios since this information gives an indication of the potential benefits of these reductions. Furthermore, we are not in a position such that we could make a better estimate of these scenarios than IIASA and nor was that the intention of this study.

Comment 7: The manuscript does include sufficient references to e.g. detailed model description papers, but the results need to be put more into context of earlier literature. The main message of the paper is to present the evolution of aerosol number concentrations; however, no attempts are made to compare the results with existing literature. While this might be the first study of aerosol number concentrations with the new IIASA emissions, there are some existing papers on future aerosol number concentrations and several papers focusing on aerosol mass with different future scenarios. The future CN and CCN concentrations are studied e.g. in Makkonen et al. (2012a, 2012b), and the presented change in aerosol number can be compared to values in those papers. The evolution of aerosol mass is studied with several different

models and scenarios, e.g. Stier et al. (2006), Kloster et al. (2008), Lamarque et al. (2011). These should at least be included in the introduction to present the reader with earlier findings of future aerosol pathways. In general, the results and discussion would benefit from several references to literature, e.g. related to how much does nucleation influence CN and CCN concentrations.

Response: We agree that these papers should be mentioned and we now discuss these references in the introduction of the updated manuscript. It is hard to compare our results with the specific numbers in these references though, since those are based on global simulations and those that focus on particle number concentrations focus on the year 2100. However, we have now added some comparison in Sect. 4.1.1 with values from Makkonen et al. (2012).

Comment 8: The effect of nucleation in number concentrations (Figure 6) seems extremely small compared to other model studies, For example Merikanto et al. (2009) find the contribution from nucleation ranging from 20 to 80% in Europe (for present-day conditions). As the scale at least in Fig.6a is huge, maybe a non-linear color-scale could be used? This would make it easier to find the effect in western and northern Europe.

Response: We have added subplots with logarithmic colour bars for Ntot and N10 to Fig. 6. What is plotted in this figure is the average factional increase in number concentration due to nucleation, defined as in Fountoukis et al. (2012) as f = (NN-N0)/N0, where NN and N0 are the number concentrations with nucleation turned on and off, respectively. The lowest continental values on f in Fig. 6a is approximately 1 (over France), or 0 in the new Fig. 6d since log10(1) = 0. An f value of 1 implies that NN is twice the value of N0, which is equivalent to that nucleation contributes 50% to the total particle number concentration. Thus, the contribution over Europe in May is in general predicted to be higher than 50%, except over northern Scandinavia.

Comment 9: The model description should include more details on aerosol dynamics,

although the reader is referred to Fountoukis et al. (2012). As the manuscript presents CCN (N100) concentrations, the role of organic vapours for particle growth should be addressed (Riipinen et al. 2011). It is mentioned in Fountoukis et al. (2012) and in this manuscript that the model underestimates the growth by organic vapours. This explains at least partly why the sensitivity of N100 to nucleation is much lower than in many other studies. If possible, this should be accounted for with an additional simulation with increased nuclei/particle growth rate due to organics. This modification would have an effect on present-day and future sensitivity of number concentrations to anthropogenic emissions.

Response: We have added a few more details on the aerosol representation in PMCAMx-UF. SOA formation does not contribute to ultrafine particle condensational growth in the current version of PMCAMx-UF, which we have clarified in the updated manuscript. We have also clarified the consequences of this in Sect. 2.

Comment 10: I feel that the Results and discussion could be sectioned more clearly, with respect to related figures. Now Section 4.1 focuses first on Ntot (Fig.1 a-d, Fig.2 a-c, Fig.3 a-d), and then moves on to N100 in the middle of section. Maybe divide this into subsections for Ntot/N10 and N100? As figures 1 and 3 present the same thing but as mean and median, I think they could be next to each other for clarity.

Response: We have added subsections to Sect. 4 and restructured the section according to what the reviewer suggested. However, we have kept the figures in the original order, since that is the order they are first mentioned.

Comment 11: Section 4.2: do the primary emissions include a diurnal cycle in the model? This is an important detail when analyzing the diurnal cycles in Fig. 4.

Response: Yes, the emissions have a one hour time resolution which is stated in Sect 3.2.

Comment 12: Figure 2: adjust the color scale symmetric around zero.

Response: We tested that, but the color scale we use now looks better than other alternatives. If the green color is to represent zero the scenario maps will be dominated by blue color which makes it more difficult to separate for instance 50% from 90% reductions. We would therefore prefer to keep the figure color scale as it is.

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