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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.

Anonymous Referee #3

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The manuscript by Ahlm et al. presents several scenarios of aerosol number concentrations over Europe for the year 2030, based on recent IIASA emission inventories. Model simulations are performed with the PCAMx-UF 3D chemical transport model, which includes detailed description of aerosols microphysics. As aerosols and especially their number concentration are important for human health and climate, the study in question is an important source of information for future predictions. It allows to estimate the effect of pollution reduction measures, even separately for several emitted compounds.

The manuscript is generally well written and contains significant new results. How-

ever, manuscript is missing several details which help to understand the generality and applicability of the results.

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.

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 can not 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.

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 PCAMx-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.

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.

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?

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?

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.

The effect of nucleation in number concentrations (Figure 6) seems extremely small compared to other model studies. E.g. 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.

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.

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.

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.

Figure 2: adjust the color scale symmetric around zero.

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