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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Primary versus secondary contributions to particle number concentrations in the European boundary layer" *by* C. L. Reddington et al.

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The paper from Reddington et al presents a very large and nice (!) compilation of data and its comparison to the GLOMAP model. Performing also several sensitivity experiments. It is very interesting to see the comparison to data done in such a broad way. The text is also very clear and the findings are valid. The paper should be published in ACP with minor revisions. Congratulations.

General remarks

The isolation of the importance of nucleation is a strength of this paper. However, I wonder if there should not be a more explicit paragraph in the discussions/conclusions

on other processes influencing size distributions, such as cloud processing, inhomogeneity of source size, dyamics of semi-volatile size distributions.

Chapter 4.1.2 and further use of statistics is used to underpin the importance of nucleation for reproducing hourly evolution of number concentrations. That is of course a possible explanation for improved correlation in some sites. However, I wonder if also a random process adding ultrafine particles would improve the statistics. This suspicion enters since the baseline simulation of eg BCOC_sm has probably a very flat, damped time series of number concentration evolution, due to missing processes to create temporal fluctuations on the order of hours. I think a random adding of number would be a useful additional valid null hypothesis to compare the experiments with BL nucleation with.

Also - shouldnt one try to evaluate a normalized diurnal cycle of number concentration. Isnt there a diurnal signal from nucleation to be expected?

I find the discussion on the primary OCBC size distribution already pretty good, and the authors are applauded for that. However, some aspects on the discussion of the potential non-validity of AeroCom size assumption could be extended :

1) I believe it is worth discussing more that the spread of the primary distribution is considerably smaller in Stier et al. This has an impact on the number of particles and this is probably as large as the mode diameter chosen.

2) The assumption of rather small initial emitted sizes might be valid if the simulation of particle dynamics is captured by the time evolution and vertical mixing of aerosol particles in the global model. The length of time steps, the vertical layering and mixing in the model and the particle dynamics may help capturing the aerosol size evolution dynamics by the model even if no nucleation is included but if small primary particles are assumed. In a way the AeroCom distribution might reflect poorly parameterized particle formation through nucleation.

3) Measurements showing larger sizes in urban areas might not reflect primary emission sizes.

4) The particle dynamics are clearly a subgrid process to a global model. Without more explicit modelling on finer scale, all size assumptions are necessarily a parameterization.

5) A larger size of BC non-volatile, primary particles might be likely. But how much is that impacting the overall number size distribution, which is a result of other condensing and nucleating aerosol components?

General remarks to all figures:

Since one emphasis of the paper is to see how different sensitivity experiments reproduce the different observed size properties, something pretty dense in information, it would be much more digestable if each experiment is identifiable with the same colour throughout all figures.

I would also like to request that a legend, eg with colored experiments names is added to all figures. It would be so much more readable if experiment abbreviations and associated color (now even changing from graph to graph) are not "hidden" in the figure caption.

Specific remarks

p 18268, l 24: "the modelled size distribution looks very different than the primary distribution"

This is probably right, but it is hard to see in the figures. The size distributions in figure 2 and 3 are plotted quite differently. Please add a characteristic size distribution (mean over all stations from "reference" simulation?) to figure 2. I think that would be illustrative and easy to do.

p 18270, l 4 : "decrease in the spatial correlation between the model with BL nucleation

and observations (BCOC lg, 0.12–0.52; BCOC sm, 0.48–0.68)"

The information in the brackets is hard to understand, since you refer not only to BCOC_lg/sm experiments, but to all experiments, right?

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 18249, 2011.