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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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Review of "Primary versus secondary contributions to particle number concentrations in the European boundary layer" by Reddington et al.

In this paper, the authors use a global aerosol microphysics model and the network of EUCAARI observations to try to deduce information about the relative importance of primary emissions and nucleation on particle concentrations (of various size classes) in Europe. It is terrific to use this large data set for model testing and improvement. The paper is well written and is certainly within the scope of ACP. It deserves to be published in ACP once several minor comments have been address.

General comments:

1.The insensitivity of particles larger than 50 nm (and particularly particles larger than 100 nm) to BL nucleation in these results is pretty remarkable. In Pierce and Adams (2009) we discuss the dampened response of CN to nucleation (this wasn't anything terribly new then, we all are aware of it), but the extent here is pretty amazing (e.g. you are shutting BL nucleation off entirely, not just scaling it by 1-2 orders of magnitude!). I believe that this could use further discussion and potentially further analysis. Obviously the dampening comes from the reduction in survival probability of ultrafine particles due to an increase in the condensation and coagulation sinks once nucleation is turned on (e.g. Pierce and Adams, 2007, Kuang et al. 2009 and numerous studies out of Kulmala's group), but it would be good to discuss this and/or look into this some more.

Pierce, J. R. and Adams, P. J.: Efficiency of cloud condensation nuclei formation from ultrafine particles, Atmos. Chem. Phys., 7, 1367–1379, 2007, http://www.atmos-chem-phys.net/7/1367/2007/.

Kuang, C., McMurry, P.H., and McCormick, A.V., Determination of cloud condensation nuclei production from measured new particle formation events, Geophys. Res. Lett., 36, L09822, doi:10.1029/2009GL037584, 2009.

2.When comparing the measured and modelled timeseries (e.g. Table 6), I believe it makes more sense to do a $\sim$5-20 hour running average of the measurements (adjust the time to the average residence time of air in the box). There will be noise on the hourly timescale of the measurements that the model cannot capture because of the spatial resolution. This would make a more apples-to-apples comparison, and I believe that comparing the r^2 values from this analysis between different model simulations would then be more meaningful.

3.There is much discussion about the appropriate size of primary particle emissions to be used for global models because the mode size increases within the first several-hundred kms from emission. This discussion is very justified, is a big problem for

global models, and it is great that this paper performs sensitivity tests to determine how primary particle size affects the comparisons to observations. I too have struggled with how to account for these issues (Pierce, J.R., Theodoritsi, G., Adams, P.J., Pandis, S.N., Parameterization of the effect of sub-grid scale aerosol dynamics on aerosol number emission rates, Journal of Aerosol Science, 40, 385-393, 2009.)

However, I do not necessarily agree with the level of favoritism of the large particle emissions in the text (e.g. P18279L9-11: "more appropriate for a global model", the numbered list on P18279L13-16 that does not include the possibility that smaller particles might be ok for the comparisons, P18286L8: "too small to be appropriate for global grid boxes"). While I agree it makes sense that the average sizes of primary particles in a gridbox must be larger than the primary particles very close to their emission, emissions can be occurring in many locations in a grid box and at various distances from the measurement locations. Therefore, the time-averaged data from any measurement site is representing some average processing time from the closest (or most influential) sources. The average processing times will be different from site to site (and they will certainly change with time/wind direction). In other words, the grid-box mean will generally not correspond to what is being measured.

These measurement-location-specific issues are difficult (impossible?) to capture in the global model. However, we need to understand that many measurement sites might normally be seeing less processed aerosol than others and that using smaller sized particles for emissions will give better agreement for these sites. It would be good to add discussion on this to the text.

Specific comments:

P18267L12-19: If you have 2 externally mixed populations, why not put all nucleated particles into one population and all primary emitted particles (BC-OC-sea salt...) into another. Allow sulfate and SOA to condense onto both. When particles from the 2 populations coagulate with each other, put the resultant particle into whichever population

had the LARGER parent particle. This formulation would give the exact contribution to CCN from nucleation versus primary emissions. You could present exact estimates of this rather than trying to tease out the influence of nucleation on CCN. I don't understand why you would put nucleated particles and sea salt into the same bins and why coagulation between particles in the two populations would always go into the same population (rather than whichever population had the bigger parent particle).

Section 4.1.2: I don't understand how the "primary aerosol" stats and "BL nucleation" stats are aggregated together when you have many different model simulations. Can you please explain more clearly?

P18282L17: "In particular, we neglect the impact of cloud cover on incoming radiation on OH concentrations". Really? Is this because you are running in a mode where OH is offline and monthly averaged? I find it hard to believe that TOMCAT doesn't predict these online.

P18285L10-14: Spacklen et al. (2006) and Hyytiala data. When I was reading the section on timeseries, I kept scratching my head thinking, "I thought BL nucleation in GLOMAP does great in Hyytiala", so I'm glad this was mentioned here. I would appreciate any additional insight into this... have things in the GLOMAP changed greatly since then? Is there a major difference between the seasons tested in Spracklen 2006 and this paper?

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