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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol ageing in an urban plume – implications for climate and health" *by* P. Roldin et al.

Anonymous Referee #1

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Review of "Aerosol ageing in an urban plume – implications for climate and health" by Roldin et al.

This paper presents an application of the ADCHEM air-trajectory model with online chemistry, aerosol microphysics and radiative transfer. The authors investigate the aging of air masses after they pass over Malmö, Sweden, and model predictions are tested using a field station 50 km downwind of Malmö. They estimate the effect of Malmö on the aerosol radiative forcing and a proxy for aerosol health effects.

The paper is a nice application of what can be done with a very detailed trajectory model, and the results have important implications. I recommend it be published in ACP once several issues and comments have been addressed.

Specific comments

1.Page 18734, line 9: 1.5 – 2500 nm. I assume this is diameter, but some groups use radius. Please specify.

2.Page 18739, 1st pagragraph: How is SOA condensation done? I could not find the following details in the other Roldin paper on ACPD. Do you track the 11x16 2D-VBS surrogate species in every size bin, or do you just track the total mass of the 11x16 2D-VBS species aerosol organic phase without size information? If you do track the organic composition at each size, do you solve condensation kinetically by solving the diffusion equation for each aerosol size section and each 2D volatility bin, or do you just assume that each size instantaneously goes to equilibrium with the gas phase? Full kinetic condensation seems very computationally demanding for 11x16 2D volatility bins, 200 size sections and 20x20 gridboxes considering the range of volatilities make condensation a very stiff system of differential equations. However, kinetic condensation may predict very different size-dependent growth rates due to SOA condensation than assuming instantaneous equilibrium or just tracking the bulk SOA mass (see comment #7). This is a tricky problem. If you are performing full kinetic condensation of the 2D-VBS onto each size section, please give details because this would be very useful.

3.Page 18741, bullet #6: There are only 5 criteria listed before this, not 6.

4.Section 2.5: This section was, in general, confusing. I deduced that you are scaling the aerosol emissions horizontally using a NOx-emission horizontal profile; however, this is never explicitly stated. Also, I believe that c_traffic should be in the numerator of the second term on the right-hand side of equation 1. Right now the units don't work out and the equation doesn't make physical sense to me. Also, I believe that the first term on the right-hand side of equation 2 should have c_j,1 rather than c_j,i-1 because it seems like you are doing a linear interpolation between the southern boarder of Malmö and the measurement station (though this was not explicitly stated, so I could

be wrong).

5.Page 18743, line 28: The criteria for finding cases where Malmö influences Vavihill is when the model-predicted number, surface area and volume all agree with the Vavihill measurements by within 10% for cases that have chemistry turned off (just dilution affecting concentrations or is there aerosol microphysics too?). Since you find cases that have close agreement to measurements without all processes turned out, it makes sense that your model might not agree as well once you turn these processes back on. Table 2 shows differences between model predictions and measurements at Vavihill that are, on average, much larger than 10% for surface area and volume. The differences are discussed in the text at the end of Section 3.1, but no discussion is made about the fact that errors were < 10% for surface area and volume when chemistry was turned off.

First, In general it is very hard to predict the entire size distribution correctly, so I am surprised that 26/232 cases actually were with 10% for number, surface area and mass (especially without chemistry. (Was this actually the criteria used for the screening, or am I misunderstanding what was written?) Second, it seems odd to start with a situation where the model gives an excellent prediction with many processes turned off, and add more processes that you know will make the model perform worse in order to address the impacts of these processes on climate and health. Third, the impact of turning on chemistry is likely that secondary aerosol mass will be increased. It makes sense that number stays in good agreement but surface area and mass do not. This will bias your climate and health-effect results.

6.Section 3.2: What about uncertainties in emissions, SOA, nucleation schemes?

7.Page 18752, line 12-14: The details of how SOA condensation is done (see comment #2) may have implications into your organic mass size distribution. If SOA is low volatility (C* $\sim<$ 1E-1 or 1E-2), it will condense to the Fuchs-corrected aerosol surface-area distribution and will not re-evaporate on a timescale shorter than what you are

modelling. If SOA is more semi-volatile, individual molecules will re-condense and re-evaporate quickly and the net condensation will be proportional to the aerosol mass distribution. This means that semi-volatile SOA will net-condense onto larger sizes than low-volatility SOA. Depending on how condensation is done (and what volatilities your SOA has if you condense using a fully kinetic condensation scheme) you can get large variability in the predicted organic size distribution.

8.Section 3.4: There seem to be many assumed parameters in the cloud radiative forcing calculation (more than just cloud depth) such as cloud fraction (what was assumed for this? are you using the cloud predictions that are included in your meteorological inputs?) and updraft velocity. How constrained are your estimates of optical depth here?

9.Did you filter out rainy days? I didn't see any discussion of the effect of these on the results. I guess it would be hard to get the number, surface area and volume distributions right on these days, which would filter them out.

10.Page 18755, line 1 and Figure 10: Are panels (a) and (c) in Figure 10 for simulations including both primary and secondary aerosols, or is it just including primary aerosols? I had assume it was both, but the last sentence on Page 18754 (that ends on page 18754) made me think it might be just primary aerosols.

Grammar comments

1.Please say "downwind of Malmö" and "upwind of Malmö" as opposed to "downwind Malmö" and "upwind Malmö". This struck me as awkward throughout the paper. To make sure I wasn't crazy I Google searched for "downwind of Copenhagen" and got 600 hits. I then search for "downwind Copenhagen" and got 4 hits (three of which were from the abstract of this paper http://www.atmos-chem-phys-discuss.net/10/8553/2010/acpd-10-8553-2010.html).

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 18731, 2010.