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Atmospheric
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Discussions

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

**P. Roldin et al.**

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We thank the referee for giving us the motivation to change the way that the model treats gas to particle partitioning and to change the way we choose our model cases. We are also thankful for the important comments, which have helped to clarify confusing or missing information in the manuscript.

We would like to clarify which changes in the model that we have performed after considering the comments from referee #1 and referee #2 as well as the comments on the companion paper (Roldin et al., 2010). The changes that concern the organic gas to particle partitioning are rewritten in chapter 2.2.2 and 2.4 in Roldin et al., 2010 (the model description paper). The description is also found in the separate document called: "Gas to particle partitioning of organic compounds in ADCHEM".

1) The 2D-VBS model has been changed and it now tracks all the 2D-VBS surrogate species in every size bin. Running this model with 200 size bins is however computationally expensive and we therefore changed to 100 size bins between 1.5 and 2500 nm in diameter. We have also decreased the number of 2D-VBS bins from 11X16 to 11X10. This is possible since we no longer consider the non-oxidized SOA-precursors in the VBS (e.g. benzene, toluene and xylene) but only their generally less volatile oxidation products. The volatility distribution in the VBS is now between C*=10-2 and C*=107 $\mu$g/m3 instead of C*=10-4 and C*=1011 $\mu$g/m3 which was the range we used for the model runs presented so far.

2) The 2-product model has been changed. It no longer uses the 2-product yield parameterizations from experimental work in smog chambers. Instead, it keeps track of each 2-product model surrogate species (2 products for each oxidation reaction). In total this gives 37 surrogate species (35 for SOA and 2 for POA). In the same way as for 2D-VBS model, the 2-product model keeps track of all 37 species in each particle size bin. However, since the number of organic compounds is fewer (37 instead of 11X10=110) this model is faster. Another advantage is that each of these 37 products can be traced back to the original non-oxidized molecules and therefore it is possible to distinguish between ASOA, BSOA and POA. This is not possible with the 2D-VBS which keeps track of the O/C-ratio (an indirect measure of the origin of the organic compounds).

Owing to these changes and the additional comment (page 18743, line 28) dealing with how we choose model cases, we are currently re-running the model for all possible cases.

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

Yes it should be diameter. We will specify it in the text.

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 2DVBS 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.

*For the results presented in the ACPD manuscript we did not track the surrogate species in every size bin, just the total mass of every condensing species. However after reading the comments by the referee we decided to update the model and now it tracks all 2D-VBS species in each size bin (see previous page). The referee points out that this is computationally more demanding especially for the coagulation algorithm. Therefore we have decided to only use 100 size bins between 1.5 and 2500 nm in diameter. As a consequence the model performs with approximately the same speed as previously with 200 size bins. With the new model version, it is only the first step oxidation products from the SOA-precursors (e.g. benzene, toluene and xylene) which are included in the 2D-VBS and not the more volatile non-oxidized compounds. With this change it was possible to decrease the number of 2D-VBS bins from 11X16 (176) to 11X10 (110) with C* between 10-2 to 107 $\mu$g/m3 instead of 10-4 to 1011 $\mu$g/m3 (See the document: “Gas to particle partitioning of organic compounds in ADCHEM”).

**The condensation/evaporation is solved kinetically in our model. However, we nor-

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mally use the wording "diffusional limited condensation/evaporation" to describe this. Hence, we understand how the referee could have missed this information. In order to avoid confusion, we have added the term "kinetic" to describe the condensation.

We have not previously tracked the organic compounds in each size bin. But, we have updated our model for this purpose. Since, we have additionally changed other model descriptions as suggested by other referee comments, we needed to rewrite several sections in our companion paper (Roldin et al., 2010) describing the model in detail. The updated version of the model description for the companion manuscript is found in the separate document: "Gas to particle partitioning of organic compounds in ADCHEM".

*** Yes we agree and we hope we have illustrated this with the updated model description.

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

Yes, thank you! We will change this error.

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).

*Yes we agree and we will rewrite the text to make it easier to understand.

**We think we have confused the referee by stating that ctraffic(Dp) means the "estimated local emission contribution". We have changed the parameter description to:

“estimated local particle number contribution”. With this in mind, the units should agree and make perfect sense. The quotient is just a dimensionless ratio multiplied with ctraffic(Dp), which is the estimated local particle number concentration contribution from road traffic with the same unit as cbackg(Dp) (#/m3).

***We understand that the referee does not fully comprehend our intention here. The procedure is very difficult to explain. The following explanation will be inserted in the model description of the manuscript. We allow for dynamic and chemical processing of the aerosol size distribution in each time step between the southern border and the measurement station at the northern part of Malmö. Hence, we cannot use a simple linear interpolation of the size distribution between these two points. We do however perform a linear interpolation for each time step, but of a different kind that includes the effects of dynamic and chemical processing on the size distribution in the previous time step. In other words, for each of the time steps (i), the interpolation is performed between the concentration one time step backwards (i-1) from the current time step and the final time step at the measurement station (time step N).

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.

*Yes, the criteria was that both number, surface area and volume for particles between 10 and 600 nm should agree within 10 % between the simplified model results and the measurements. We should clarify in the article that it is the particle concentrations for particles between 10 and 600 nm which were used as criteria and not the total concentrations between 1.5 and 2500 nm. This simplified model did include vertical mixing, all aerosol dynamic processes (except homogeneous nucleation) including condensation, but using a fixed concentration of condensable compounds (107 molec/cm3, zero saturation vapor pressure and molar mass of 150 g/mol). We agree that taking into account the limitations with this model we should use wider criteria for number, surface area and volume. We have gone through all 232 simplified model results again and changed the criteria to allow 20 % difference for number, particle surface area and 30 % difference for volume. We then find 67 cases which obey these criteria. Although this is a higher number than previously, all cases are however from the same 26 days as before. Nevertheless, the increased number of cases will help us make better selection of final cases as follows: We are currently running the complex ADCHEM model with detailed gas and particle phase chemistry in 1D. From these model runs we will select the final cases, with the requirement that the number, surface area and volume should agree within 10 % with the measurements. If more than 2 cases for the same day show this good agreement, the best of these cases is selected. Hence, we will in the end anyway have a maximum of 26 cases or possibly less, but the selected best case for each day might change a few hours in time of the day.

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** Yes we agree. For this reason we think it is important to select more cases from the 232 than previously and model them with a more complex model and afterwards select the final cases which should be used (see *).

The difference in the urban contribution for surface area and volume is indeed larger than 10 % as the referee correctly points out. However, the urban contribution should not be confused with the measured concentration at Vavihill. The average and median number, surface area and volume concentration agrees within 10% between the model and measurements (3 % for mean number, 0.2 % for median number, 4 % for mean surface area, 5 % for median surface area, 6 % for mean volume and 9 % for median volume). Since the written explanation and the corresponding table seem to confuse the readers we will make a better distinction between the urban contribution and the total concentration in the updated version of the manuscript.

***The referee asks the question whether he has understood correctly the selection criteria? The answer is: Yes. The referee also raises a concern that the narrow selection criteria for the volume concentration could produce bias for the final results in terms of secondary aerosol mass. With the old criteria this could indeed possibly be the case. However, with the new selection criteria allowing for larger variability in volume concentrations, we don’t rely as much on the simplified model runs as previously, and hence, this possible bias is suppressed (see *).

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

There are indeed many different uncertainties affecting the model performance. The large uncertainty in the homogeneous nucleation rate and initial growth rate of these particles is one of them. However, for none of the 26 days which were modeled, significant new particle formation was occurring for several consecutive hours at both measurement stations. This indicates that for none of these days large scale regional nucleation events were occurring. Hence, the new particle formation between Malmö and Vavihill were likely also moderate or insignificant for these days, indicated by the good agreement between the model and the measurements at Vavihill. For 2 out of 26 days large numbers of particles were however found in the nucleation mode at the measurement station in Malmö. These particles were likely formed from local sources in or near Malmö (e.g. ship traffic). Even though none of the 26 selected days were characterized by large scale nucleation events, homogeneous nucleation can possibly have biased the average results presented in this study. This is since the simplified model used to make a first selection of possible days with influence from Malmö at the Vavihill station did not consider homogeneous nucleation and therefore systematically filtered out days with large new particle formation between Malmö and Vavihill. Further, primary particle emissions downwind of Malmö are uncertain. Although only marginal effects are expected as an effect of this uncertainty between Malmö and Vavihill, the uncertainty might become much larger up to 24 hours downwind of Malmö. The gaseous emissions both upwind and downwind of Malmö are also uncertain (especially for the IVOCs), which affects the precision of secondary aerosol formation. Finally, most of the reactions involved in the secondary organic aerosol formation are unknown, which gives high uncertainty for this process. We will add an extended discussion of these uncertainties and other effects related to these uncertainties in the revised version of the manuscript.

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* _< 1E-1 or 1E-2), it will condense to the Fuchs-corrected aerosol surfacearea 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 reevaporate 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.

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Yes, thank you for describing this clearly to us. This is indeed very interesting and important to mention in the text. This is a large advantage of the updated model which keeps track of each 2D-VBS compound in each size bin and in the gas phase and then solves the condensation/evaporation separately for each of these compounds (See the document: "Gas to particle partitioning of organic compounds in ADCHEM"). We will add a comment about this in the final manuscript version and illustrate how the 2D-VBS compounds distribute differently into different size bins depending on their volatility.

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?

*Yes, we need to point out clearly that these are just examples of how the radiative forcing would become during completely cloudy days (cloud fraction=1). We are not aiming at giving the best estimate of the yearly average radiative forcing induced by Malmö's contributions to the cloud properties, but just illustrate that the effect of the emissions can be significantly different during cloudy and non-cloudy days. We are going to clarify this better in the text.

** No, these estimated cases are just hypothetical assuming that we have low level clouds positioned at the top of the boundary layer which are formed from adiabatically ricing air-parcels fed by the particle properties at the ground.

***We use an updraft velocity of 1 m/s. According to Rogers and Yau, 1989, typical updraft velocities for stratus clouds are on the order of a few tens of centimeter per second and for cumulus clouds in the order of meters per second. We will add this reference to the text. ****We don't know since we do not compare the model results with actual measurements of the cloud optical depth. We will provide information about the optical depth of the modeled clouds and will carefully consider how we use the term

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“optically thick clouds”.

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.

Yes, we selected days without rainfall between Malmö and Vavihill along the HYSPLIT trajectories. However downwind of Vavihill and upwind of Malmö rainfall was occurring for several of the simulations. We will clarify this in the text.

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.

Panel (a) and (b) in figure 10 are the results from simulations with both primary and secondary aerosols. In figure (c) and (d) we illustrate the secondary aerosol contribution to the radiative forcing. This is calculated as the difference between the radiative forcing when including both primary and secondary aerosols (base case model runs) and the radiative forcing of primary aerosols only (no anthropogenic gas phase emissions in Malmö).This will be clarified better in the figure caption.

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-physdiscuss. net/10/8553/2010/acpd-10-8553-2010.html).

Yes, thank you! We will change to downwind of Malmö and upwind of Malmö.

References

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Press, Oxford, Great Britain ISBN: 0-08-034864-5, 1989.

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Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/10/C9901/2010/acpd-10-C9901-2010-supplement.pdf

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