We appreciate the comments by the referee which have helped us to in a better way understand the uncertainties related to the meteorological input data, air mass trajectories and the vertical and horizontal dispersion.

We would like to clarify which changes in the model that we have performed especially after considering the comments from referee #1 as well as the comments on the companion paper (Roldin et al., 2010). The changes concern the organic gas to particle partitioning which is described in Sect. 2.2.2 and 2.4 and new model results from 2D-VBS and two-product model simulations are presented in Sect. 4.2.1.

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 $C^* = 10^7 \mu g/m^3$ instead of $C^* = 10^{-4}$ and $C^* = 10^{11} \mu g/m^3$ 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 40 surrogate species (35 for SOA, 2 for POA non-oxidized POA, 2 for non-oxidized IVOCs and 1 for oxidized POA and IVOC products). In the same way as for 2D-VBS model, the 2-product model keeps track of all 40 species in each particle size bin. However, since the number of organic compounds is fewer (40 instead of 11x10=110) this model is faster. Another advantage is that each of these 40 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).

1. In several places, the authors mentioned that the model is developed for detailed studies from local scale to regional or global scale. ADCHEM is a Lagrangian model and air masses in different grid boxes may have quite different trajectories. As I understand, the model follows one single trajectory for the whole domain and it is not clear how accurate the results will be after a few days of air mass travel. The authors need to explain why the model can be used for regional or global scale.

Yes, thank you for addressing this issue. We will not state that ADCHEM should be used for global scale in the updated manuscript. We will include a discussion about the
scales which ADCHEM is appropriate for and under which meteorological conditions the assumption of following one air mass trajectory is not suitable.

2. Model input (Section 3.2). This session should be expanded to provide more details on how vertical profiles of meteorology parameters are obtained and used to drive the model. Could the authors provide a figure showing such profiles? Do these parameters change horizontally? Since the vertical profiles are used, I assume that wind speed at least vary with height. Then, different air masses in the domain travelled different distance during a given period of time. How do you deal with this in the model? What about the wind shear? Have you considered the effect of wind shear on mixing? The authors refer to Roldin et al. (2010) for more information but I could not find much of such information there either. Anyway, this one is the model description paper and it is necessary to provide such information here.

Yes, we agree that we should include a more detailed description about the meteorological data and how it is used in the model. We will do this in the updated manuscript. We will include one figure in the supplementary material which gives examples of temperature and relative humidity profiles at a few places along the air mass trajectory downwind of Malmö. We will also clarify in the text that the meteorological conditions which we use do not change horizontally perpendicular to the air mass trajectory which ADCHEM follows. We will also include a figure in the supplementary material which illustrate how different HYSPLIT air mass trajectories which start at different altitudes over Malmö are diverged from each other due to wind shear. We will also clearly clarify that since ADCHEM is a Lagrange model it only follows one air mass trajectory and does not consider the wind shear induced divergence of the air mass trajectories at different altitudes. This is an important disadvantage with Lagrange models compared to Eulerian models. ADCHEM do however consider how the wind shear and buoyant convection influenced the turbulent diffusivity in the horizontal and vertical direction. We will clarify this in the updated manuscript.

3. Compared to 0-D Lagrangian box-models, one advantage of the ADCHEM model is its 2-D spatial distributions. The figures presented show the vertical spatial variations but no figure was given to show the horizontal variations. I think that it will be useful to demonstrate the capability of the ADCHEM model by presenting one or two figures showing the horizontal spatial variations of key species. One good example will be the concentrations of particles in the whole domain (2-D vertical-horizontal cross section) at selected times (or locations).

Yes, certainly it would be good to demonstrate how the particle number concentration changes in the horizontal and vertical direction perpendicular to the air mass trajectory. We will include such a figure displaying the particle number concentration within the whole 2D-model domain at a few different distances downwind of Malmö. The authors stated that “the particle number size distribution in the center of the urban plume from Malmö is mainly affected by dry deposition, coagulation and condensation”. What about nucleation? Nucleation is well known to occur frequently in Europe and is a key process controlling particle number concentration. It appears that nucleation was not important for the case study presented here. Have the authors looked into cases where significant nucleation occurred? If yes, how well was the modeling doing for such cases?

No for the urban plume studies performed with the model we have not considered any cases with significant homogeneous new particle formation. The maximum new particle formation rate for the selected case study was approximately 1 cm\(^{-3}\) of 1.5 nm stable clusters. Still, these particles did not have any significant influence on the particle number concentration for particle larger than 5 nm in diameters. This is likely since the growth rate of these particles was too low for them to escape the coagulation sink. We will add a discussion about this in the text where we will state that the uncertainty concerning the influence from homogeneous nucleation is especially large since the growth rate of these particles depend strongly on the concentrations of low-volatile organic compounds which we do not know the concentration of. Therefore it is possible that ADCHEM significantly underestimated the growth rate of the smallest particles.
5. Page 18668, line 23 and below. Could you explain in more detail how this scheme (usage of an inert specie) is related to the diffusion of model tracers?

Yes, we will clarify this in the manuscript. The use of an inert specie is possible since the transport of compounds by turbulent diffusivity in contrast to e.g. molecular diffusion is independent of the properties of the individual species. The mixing of the inert specie therefore describes the air mass transport between different grid cells. Since we use operator splitting and first consider the mixing and then the species specific processes (e.g. coagulation, dry deposition and chemical reactions) the general air mass transport between the grid cells is the only information which is required.

6. Page 18668, lines 11-14. How sensitive is the result to assumed concentration gradient values in the upper boundary? How you treat the mixing around the horizontal boundary?

Yes thank you for this important question. We have also run the model without any concentration gradient as upper boundary conditions. However since the atmosphere above the boundary layer is stable the mixing in the vertical direction is very slow. Therefore this had insignificant influence on the concentrations within the boundary layer. Near the upper boundary conditions the concentrations do however become higher if using a zero gradient. Unfortunately we have no measurements on gas or particle concentrations in the vertical direction for the studied case study. We will include a discussion about this in the updated manuscript.

As horizontal boundary condition we assume that the concentration gradient is zero. We will clearly write this in the updated manuscript.

7. Page 18676, line 18. It appears that the output from HYSPLIT model is used heavily in the ADCHEM model. It is necessary to give a reasonable description of the HYSPLIT model and its uncertainties.

Yes you are right. Since the HYSPLIT model rely on relatively coarse meteorological input data (1°x1°) the local wind field may differ considerably from what is predicted by the HYSPLIT model. Therefore we used wind direction measurements at 24 ma.g.l. from a meteorological mast in Malmö to verify the HYSPLIT air mass trajectory. For the selected case study the air mass trajectory agreed within 5° with these measurements. We will add this information to the manuscript. Unfortunately, we have not been able to find any document concerning the model uncertainties with the HYSPLIT model. We will however referee in the text to the comprehensive trajectory model intercomparison by Stohl et al., 2001 which compared three different air mass trajectory models and concluded that close to the surface the average horizontal position deviations may be up to 10% for 48 hour backward trajectory simulations.

References


Interactive comment on Atmos. Chem. Phys. Discuss., 10, 18661, 2010.