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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.

Anonymous Referee #2

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In their paper, Roldin et al. use a model for aerosol dynamics, gas phase chemistry and radiative transfer calculations (ADCHEM) to estimate the influence of the urban particle and gas phase emissions in Malmo, on climate and health relevant particle properties downwind the city. The paper is, in general, well written, and contains relevant scientific information that will add to the current level of understanding of the ageing of urban emissions, from small to midsize cities, from urban background to regional scales. I recommend the paper for final publication, after some issues are addressed.

Specific comments:

1. Section 2.1 Page 18736: The ToF-AMS measurements were used to test the accuracy of the ADCHEM model in describing the aerosol chemical composition. As these measurements were not conducted at the time of the modeled period the authors should also mention which was the simulation period and also comment on the expected uncertainties by evaluating the model with these measurements (especially if the simulations was not during the October and March). In addition, it is not clear whether the gas phase concentration measurements were held during the simulation period.

2. Section 2.2 Page 18737: The authors provide a brief description of the different schemes that the model can use to simulate the aerosol dynamics, the SOA formation, and the condensation/evaporation of the inorganic components. However, there are no indications of how these different schemes can affect the results and why the methods described in the last paragraph of this section (Page 18739, lines 1-8) were selected to serve as basecase. An analysis of the sensitivity of the results to these assumptions would be appropriate along with a more detailed discussion for the selected schemes (i.e. the uncoupled condensation of acids was chosen. Are there any indications that the particles during the simulations are not acid neutralize?)

3. Page 18737 2nd Paragraph: It is not clear how the model treats the organic aerosols coming from the oxidation of the Semi-volatile POA and the intermediate VOC when the 2D-VBS is used. Are they lumped with the ASOA? Moreover, it would be helpful to provide the ageing reaction rate and an explanation of the way that the organic aerosols shift their position in the 2D matrix after each oxidation (what’s the depletion on their volatility? What’s the increase on the O/C ratio?).

4. Page 18738 line 6: Please provide a reference for the chemical mechanism used.

5. Page 18738 lines 14-16: How the high NOx/HO2 is defined? Moreover, depending on the value of the NOx/HO2 ratio, does the model follow one of the two reaction pathways (NO vs HO2) or a combination of those two? I would also recommend to provide a table with the SOA-yields used under the high and low NOx/HO2 ratios.

6. Section 2.7: I found this exercise very interesting. Based on these results, instead of using the measured size distribution at Vavihill as representative upwind Malmo

conditions, you could use 8% higher particle number concentrations than the measured background values at Vavihill.

7. Section 2.8: The authors should give more information for the simulation run without the anthropogenic gas emissions. In that case, are POA emissions treated as non-volatile? This would probably introduce an error in the estimated effect of the secondary aerosols as the concentration of POA would be increased. If the 2D-VBS theory is still used, how exactly the oxidation products of the low and semi-volatile organic compounds are being excluded? Part of OA is emitted and remains in the aerosol phase for its entire atmospheric life and therefore should be considered POA. Another part is emitted in the aerosol phase but it can evaporate in lower OA conditions and oxidized during its trip to Vavihill and re-condense in the aerosol phase forming SOA. Probably the correct way would be to allow the gas-particle partitioning of OA but ignore the ageing reactions.

8. Page 18746 line 15: Please provide reference for the updraft velocity. In addition the authors should provide information for the values of the rest parameters that constrain the model results (i.e. accommodation coefficient).

9. Page 18750 lines 8-9: This sentence implies that the authors have not actually calculated the ground number concentration error. Instead, they have assumed that this error should be less than 20%. If that is the case, I suggest adding a more comprehensive discussion about what led the authors to make such an assumption and also avoid referring in arbitrary numbers. Otherwise, please re-write this sentence in order to avoid this misunderstanding.

10. Page 18752 line 19: What is the physical meaning of the negative contribution (-0.01) of SA to the total PM2.5 mass?

11. Figures 6, 7, 8: It would be interesting for these figures to display the modeled contributions of POA and SOA separately.

Technical corrections:

1. Page 18733 line 18: replace “Therefore is it important. . .” with “Therefore it is important. . .”

2. Page 18741 line 23: “. . .criteria 1 to 6. . .” should be “. . . criteria 1 to 5. . .”

3. Page 18742 Eq. (1): The Ctraffic(Dp) should be on the numerator.

4. Page 18745 line 13: insert comma after “Malmo”.

5. Page 18748 line 4: “Fig 4a and b” should be “Fig 3a and b”

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