

Interactive comment on “Simulating regional scale secondary organic aerosol formation during the TORCH 2003 campaign in the southern UK” by D. Johnson et al.

Anonymous Referee #1

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General comments:

The manuscript presents the results of a box model based on the well-documented chemical mechanism MCM3.1 and compares the results to field measurements with various environmental and pollution influences. The work presented here is original and worth to be published in Atmospheric Chemistry and Physics. However, the manuscript requires some major improvements prior its publication.

Specific comments:

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1) Section 3.2, first paragraph: The Goodwin et al. (2001) speciation for the UK (since it is based on NAEI) is valid also for the other European countries? Especially for the industrial emissions, one will expect major differences between different countries, since their industrial activity is not expected to be the same. Do you have a feeling on how much this assumption will affect the results?

2) Page 7837, equation 1: Do you expect that this relationship will be the same for non-urban locations? This is a major assumption, since POA also come from biomass burning and from the oceans, while there are also NO_x emissions from soils. By assuming that the aerosol emissions are always proportional to NO_x, when simulating rural atmosphere, you will get (low, but existent) POA emissions. Furthermore, by assuming the constant value of 0.012 you assume that e.g. cars, industries and biomass burning emit the same amount of POA for a specific amount of emitted NO_x, which is clearly not correct.

3) Page 7837, line 7: Can you say that the ratio of OC to EC of 1.1 observed in a UK city is representative for your whole studied area, including the trajectories that the model follows (i.e. a big area of NW Europe and the North Atlantic)?

4) Page 7837, lines 16-21: Is this assumption valid for the other countries?

5) Page 7838, line 15: How many emitted VOC does the CRI contains? The net VOC mass that the two models use are the same, after the appropriate lumping?

6) Page 7839, 2nd paragraph: Do you have OA measurements for this kind of atmosphere? If yes, how are these measurements compared with your value of 0.7 (source (ii) in your results)? Why not assuming an initial OA mass (or POA mass) and then use it as an initial concentration, instead of using the source (ii) as mentioned in page 7847, lines 6-7?

7) Page 7839, 2nd paragraph: Are all your trajectories start from the North Atlantic? If not, maybe you should change your initial concentrations for the trajectories, and how

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much will this affect the results?

8) Page 7840, equation 3: In the original paper by Pankow (1994), there is also a factor f in the numerator that denotes the fraction of the absorptive aerosol mass. Even if you assume that this factor is unity, you should not eliminate it and add a short comment about it. It will also affect the discussion in section 5.2.2.

9) Page 7841, equation 5: No changes with time, while the condensation of gaseous species occur. This will affect the final partitioning of the semi-volatile species. Do you calculate the partitioning iteratively, or you just calculate the equilibrium in your time-step just once? If you assume that for every time-step the gas-to-particle transfer is being done only once, then you never reach equilibrium, and you will have more mass in the gas-phase than in the aerosol-phase, which might be an additional reason for your necessity to increase the K_p values.

10) Page 7841, line 22: Is this your uncertainty for the k_{in} value?

11) Page 7843, line 29: Why do you have greater input from Utembe et al. (2005)? If there are differences between the models, they should be clearly declared.

12) Page 7844, 2nd paragraph: Since your ultimate goal is to simulate the measurements correctly, there is no reason to focus on the discrepancies between the two models, but with measurements instead. A discussion for the reasons that the two (almost similar?) models give different results is still important, but not the most important one. Additionally, if you believe that the reason of this discrepancy is the reactions mentioned and the temperature difference, these are not applicable in the Utembe et al. (2005) study?

13) Page 7845, line 28: How do you specify the "good consistency"? If you compare your results with Figure 12 of McFiggans et al. (2005) there are definitely similarities, but also differences. You can compare for example the agreement between the two studies for the end of July and 1/8, but the high ratio of HOA/OA in McFiggans et al.

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(2005) that continue to exist at 2/8 is not the case for the present study.

14) Page 7846, last paragraph: Jenkin (2004) has increased the K_p values by a factor of 120, and Johnson et al. (2004) by a factor of 20-80. How did you reach the factor of 500? Although it is an arbitrary value, the reason of increasing just to match the observations is not enough. You could have a factor of 120 instead of 500, then you would get an underestimation of the OA values and finally your conclusions would be different.

15) Page 7847, source (iii) and section 5.2: One of my main concerns is the increment of the K_p values. How do you compare the K_p values that you calculate with the estimated lumped values that Hoffmann et al. (1997) and Griffin et al. (1999) have calculated in chamber experiments, and are widely used in global modeling studies? A comparison table would be very informative. The increase by a factor of 500 is extremely high and is definitely not species-independent. Some species are more favourable to accretion reactions than others, and this rate is expected to differ with varying aerosol chemical composition and acidity. It would be very interesting to try to make a first approach on different increments of the K_p values based, say, on the species structure. Since your model is so detailed, it could be very interesting to do such a step ahead, rather than using the same (actually, higher by a factor of 4-25) scaling factor with the work previously published.

16) Page 7847, line 16: Why source (ii) is SOA and not a mixture of POA and SOA? This also applies to page 7850, line 22, where the OA background will also contain POA, from my respect, and it will not have the same C:O:H:N ratio.

17) Page 7848, line 2: The underestimation of OA for the 8/8 can be attributed to the underestimation of O₃ for the same period (Figure 1)?

18) Page 7848, 2nd paragraph: Are you sure that you have all POA sources in your model? Do you include biomass burning and an oceanic source? The "ubiquitous background concentration" is a really strange quantity, which I assume that you could

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be able to get rid of, if you include an initial POA concentration. The intercept in Figure 7 (page 7849, line 22) should be resolved in your model, like the O₃ intercept does.

19) Page 7848, last paragraph: Since the definition of non-volatile species is the ones that do not exist (mostly, if not totally) in the gas-phase, then why you have so small contribution of them to the aerosol-phase?

20) Page 7853, lines 3 and 6: Aerosol formation is not a linear process, thus you can not just make a division such as $500/2.5=200$.

21) Page 7855, line 22: If 170 is your average MW, you do you use a value of 130 in equation 6?

22) Page 7856, first lines: WSOC will not only partition with their K_p values but also with their Henry values, if you have enough water available on the particles (Griffin et al., 2003).

23) A comparison of aerosol-phase species, like in figure 2 for the gas-phase species would be very interesting, if aerosol-phase measurements are available.

Technical corrections:

1) Page 7830, line 9: the model has been developed elsewhere, and only used here (even if it has some modifications).

2) Page 7832, line 14: "representative biogenic VOC", please name them, like you do in the conclusions, page 7856, lines 24-25.

3) Page 7835, line 14: For which year are the mapped emissions of Derwent et al. (1996)?

4) Figure 1: Remove 00:00.

5) Figure 5: Gridlines are not useful, you can add the 1:2/2:1 lines or any other ratio you think as informative instead.

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References (not mentioned in the manuscript)

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