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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimating the influence of the secondary organic aerosols on present climate using ECHAM5-HAM" *by* D. O'Donnell et al.

D. O'Donnell et al.

declan.odonnell@env.ethz.ch

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We would like to thank the Reviewer for his/her thorough and careful review, which will improve this paper substantially. We reply to each of these comments below.

The manuscript could be improved by some reorganization and by more concise wording. Several global models (generally global chemical transport models) already contain SOA. Unique aspects of this work, such as calculation of AOD and direct effects or the use of the Saathoff parameterization for temperature dependent SOA should receive more attention while the description of SOA partitioning basics and emissions could receive less.

Response: As suggested in the Reviwer's specific comment 5, we will relegate the details of the gas/aerosol partitioning to the Appendix. We accept that the novel aspects of this work need to be set out with greater clarity.

1. Abstract: Much of the abstract (about the first 75%) reads more like an introduction than an abstract. Space in the abstract could be more beneficially used to provide details on the discoveries of the manuscript (more results, description of what was performed).

Response: Comment accepted. The abstract will be substantially improved in the revised manuscript.

2. In several locations, the manuscript could be more concise and have a more effective message.

Response: We accept that the manuscript needs to revised with conciseness in mind. The examples given by the Reviewer are helpful in this regard.

3. Computational limitations. Page 2412, line 26 – page 2413, line 2 indicate that oxidation reactions with OH, O3, and NO3 are taken into account, but only the major pathways are considered to produce SOA. Monoterpene+OH SOA is neglected. Table 3 indicates that the production of SOA from monoterpenes is quite low considering the magnitude of emissions used (ECHAM-HAM5 is producing about the same amount of monoterpene SOA as Heald et al whose monoterpene emissions are about half of those in this work). There are many options for lumping SOA tracers to reduce computational burden while capturing all relevant SOA pathways. For example, Chung and Seinfeld (2002) lumped monoterpene and sesquiterpene oxidation by O3, OH, and NO3 into 5 parent hydrocarbons systems. Volatility basis set approaches (such as those by Jathar et al. ACPD 2011) lump together SOA from isoprene and terpenes based on volatility. Monoterpene + OH SOA formation in ECHAM-HAM could be parameterized in such a way as to produce species whose volatility can be lumped together with the monoterpene + O3 products with little error. Since the gas-phase

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reaction is already being computed, very little additional computational burden should be incurred.

Response: The added computational burden from SOA does not arise from the chemistry parameterisation, and this is not why we chose such a limited approach. The main load increase arises from the increased number of aerosol species. We have a size-resolved model, in which each SOA species occurs in four aerosol modes. For each two-product pair, this means eight new aerosol cases that must be treated with respect to aerosol microphysics, wet deposition, and other processes, and it is mainly this that gives the substantial (35-50% over ECHAM5/HAM without SOA) CPU increase. The volatility basis set has the advantage of giving a fixed number of SOA aerosol cases irrespective of the number of precursors and SOA pathways, and we see this as the longer-term solution for SOA in ECHAM5/HAM.

4. Language. As a stylistic choice, I would reduce highly subjective language such as page 2414, line 22 that states that questions regarding SOA and aerosol water are "more intractable" than others.

Response: Comment accepted.

5. Since equations 4-10 are nothing new, perhaps they should be moved to an appendix and only the most relevant, final equation placed in the main manuscript.

Response: Comment accepted.

6. Page 2417, line 17-18: Just to clarify, Is Mo being calculated from equation (9) or is the value from another timestep being used? It seems as though you need more than eqn 7, 8, and 13.

Response: Mo is indeed calculated from (9). This will be corrected in the revised manuscript.

7. Page 2417, Line 19-25: These paragraphs should be combined and rewritten to clarify which modes actually contain SOA. The paragraph states that the model strongly

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favors the larger modes, but there is no primary organic aerosol in that mode (at least for the insoluable one) for the SOA to partition into. Later in the manuscript, the lack of primary organic aerosol in the nucleation mode is also highlighted. Thus, SOA should really only be present in a couple of the 7 modes. A succinct description of the modes with SOA (and perhaps their relative amounts) would be helpful.

Response: SOA is present in four of the seven aerosol modes, namely the insoluble Aitken mode, and the soluble Aitken, accumulation and coarse modes (the same modes that contain primary organic aerosol). One sees from (13) that the amount of SOA in any of these modes follows the mass of primary OC (as a proportion of total OC). In practice, this means that in this model, most SOA occurs in the accumulation mode. This information will be added to the manuscript.

8. Page 2419, line 22: What Henry's law parameter was used to govern scavenging of gas-phase semivolatiles?

Response: For the more volatile of the two products, we took the value of 'Generic Aldehyde' (4200) from the Sander list. For the less volatile, a value of 10e5 was taken as a representative value among polyols.

9. Page 2420, Lines 11: what model dynamics are being calculated in spectral space? Please add a reference if possible.

Response: We will add a reference to the ECHAM5 model description, published by the Max Planck Institute for Meteorology in Hamburg. The model calculates the vorticity and divergence of the wind field, temperature and the log of surface pressure in spectral space.

10. Page 2422, line 11-14: This sentence is a bit long. Consider rewriting for clarity.

Response: Comment accepted.

11. Page 2423, line 17-24: Zonal mean plots of SOA show a local minimum in the vertical structure at approximately 5 km which is primarily attributed to partitioning of

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the less volatile isoprene SOA species near the surface and the more volatile isoprene SOA species aloft due to colder temperatures. How much of this structure is due to physical processes like convection? Figure 5 indicates that without SOA, there is also a local minimum although it is much less pronounced and perhaps shifted slightly higher in altitude. Are there issues due to depletion of OH (as a result of isoprene+OH, see for example Archibald et al. 2010) that could result in isoprene being transported aloft before being oxidized? The authors should not be expected to resolve and OH depletion issues, but it would be useful to point out if it might be occurring.

Response: The structure to which the Reviewer refers is entirely driven by convection. Oxidation of isoprene and monoterpenes takes place rapidly near the surface. In the case without SOA, no oxidation chemistry is involved. In this case, we simply have two emission maxima, one in the Northern Hemisphere mainly due to fossil fuel burning, and one in the Southern Hemisphere mainly due to biomass burning, and the local minimum to which the reviewer refers is simply the intervening lower-emission zone. As regards possible OH depletion issues, OH is prescribed in the model and we assume that it is non-limiting with respect to isoprene consumption. The model gives a short isoprene lifetime (approximately 1 hour in the global mean) and isoprene concentrations of <1 ppbv in the upper troposphere.

12. Page 2424, line 9. Kp is independent of Mo but this phrasing indicates it was calculated at an Mo. Consider rewording.

Response: The comment is correct, this example will be corrected in the revised manuscript.

13. Page 2425, Section 2.6. Can more information be provided about the inputs used to calculate AOD?

Response: Nothing new is added in the model development described in in this paper, we refer the reader to the description given in the referenced paper by Stier et al. (2005) that describes the ECHAM5/HAM model.

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14. Page 2426, Line 16: Particles with a radius of 35 nm can act as CCN. Does this mean CCN activation is not a function of supersaturation?

Response: No, this sentence is intended as a statement that CCN activation is not a function of chemical composition. It will be clarified in the revised manuscript.

15. Page 2427, Section 4: As a reader, I think reading about how the model's organic aerosol concentrations compared to observations before the direct/indirect effect estimates gives greater confidence to those estimates. Consider moving the evaluation section before the direct effect calculation.

Response: While we see merit in this suggestion, it would be contrary to the chapter structure, which places all model results in chapter 3, the comparison with observations other models in chapter 4, and comments and discussion on the results and the comparisons in chapter 5. We see a restructuring as carrying too great a risk of confusing, rather than improving, the paper.

16. Page 2427, line 15- Page 2428, Line 4: The authors state that semivolatile species should be measured in situ to preserve the partitioning between the gas and aerosol phases. Although these measurements might be the most desirable, there are meaningful measurements that could be made on the ground (for example, specific chemical compound identification, gas+particle analysis).

Response: It is certainly not our intention to suggest that such measurements have no value. We state only that aerosol mass measured in such a way cannot be used for the validation of modelled semi-volatile aerosol masses.

17. Page 2428, Line 10: What OM/OC ratio was used to convert observations of OC to OM for the different networks?

Response: For EMEP, we apply a factor of 1.4 as recommended in the EMEP manual (http://tarantula.nilu.no/projects/ccc/manual/index.html). IMPROVE provides OM data.

18. Section 4.1.1 and 4.1.2: Tables showing the modeled and observed values as well

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as correlations, etc would be easier to read than the values in sentence form.

Response: Comment accepted.

19. Page 2429, Line 9: Figure 15 should be Figure 14

Response: Will be corrected in the revised manuscript.

20. Page 2434, Line 6: Please add a reference for methyl chavicol SOA.

Response: Reference will be added in the revised manuscript.

21. Page 2431, Line 1-2: The manuscript states that the high degree of correlation between EC and OC in the EMEP data indicates that the OC content is largely anthropogenic. A clarification could be added to highlight the fact that the carbon could be from a biogenic source (like isoprene from plants), but the SOA might result from an anthropogenic oxidant or other anthropogenic effect that enhances the SOA (for example, see Carlton et al. 2010). Figure 4 indicates that biogenic SOA is quite high over Southern Europe.

Response: We agree and will take this point into the revised manuscript.

22. Page 2436, Section 5.3: This paragraph on the effect of NOx on SOA could be shortened and placed at the end of page 2413.

Response: We agree that it can be shortened, but we believe that it would better merged into section 5.2, rather than into the model description part. A short sentence will be added to the model description stating the lack of NOx dependency in the model.

23. Page 2436, Line 18: optical typo

Response: Will be corrected in the revised manuscript.

24. Table 1: Add Saathoff reference to reference list

Response: Will be corrected in the revised manuscript.

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25. Table 3: Henze et al. 2008 used GEOS-Chem

Response: Will be corrected in the revised manuscript.

26. Figure 5-6: Can the number of digits on the color bar be reduced?

Response: Unfortunately not without losing the lower values altogether. We need the lower values for the monoterpene SOA. The alternative is to put the plots for isoprene and monoterpene SOA on different scales, which is not an attractive option either.

27. Stylistic comment: Several different styles of plotting are used (for example, figure 11 vs 12). A more uniform presentation would be desirable for publication.

Response: We will try to re-plot figure 12 in the style of figure 11. The zonal mean plots are unfortunately not possible to make with that software, so there is little we can do about the different styles there, but we agree that at least the surface plots should be more uniform.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 2407, 2011.