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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.

**Anonymous Referee #1**

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This paper updates the global climate model ECHAM5-HAM to include secondary organic aerosol formation from isoperene, monoterpenes, and light aromatics. The updated model is evaluated against surface organic aerosol measurements from U.S. and European networks and AOD measurements from AERONET. The model is used to estimate the radiative effect of SOA.

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.

Comments:

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).

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

For example, Page 2412, Line 24 could be removed. If desired, the authors could rephrase line 26, page 2411 to emphasize that the precursors are gas-phase by stating. . . "Gas-phase precursor species included in the model. . ."

Section 5.1 could be entirely removed and replaced by one sentence on page 2421, line 5 "Global annual totals are calculated as 446 Tg/yr isoprene and 89 Tg/yr monoterpenes, in close agreement with the estimates of XX Tg/yr and XX Tg/yr from Guenther et al. (YYYY)."

Page 2433, Line 21 through the top of the next page that highlight the fact that year 1990 speciation was used is essentially stated on page 2412. Page 2414 Lines 8-21: Could be shorter and more succinct.

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 re-



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

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.

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.

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.

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 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.

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



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

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

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 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.

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

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

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?

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.

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 mean-



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ingful measurements that could be made on the ground (for example, specific chemical compound identification, gas+particle analysis).

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

18. Section 4.1.1 and 4.1.2: Tables showing the modeled and observed values as well as correlations, etc would be easier to read than the values in sentence form.

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

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

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.

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.

23. Page 2436, Line 18: optical typo

24. Table 1: Add Saathoff reference to reference list

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

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

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.

Archibald, A. T., Jenkin, M. E., and Shallcross, D. E.: An isoprene mechanism inter-



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comparison, Atmos. Environ. 44, 5356–5364, doi:10.1016/J.ATMOSENV.2009.09.016, 2010.

Carlton, A. G., Pinder, R. W., Bhave, P. V., and Pouliot, G. A.: To what extent can biogenic SOA be controlled?, Environ. Sci. Technol., 44, 3376–3380, doi:10.1021/Es903506b, 2010.

Jathur, S. H., Farina, S. C., Robinson, A. L., and Adams, P.J.: The influence of semi-volatile and reactive primary emission on the abundance and properties of global organic aerosol, Atmos. Chem. Phys. Discuss., 2011.

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