

# ***Interactive comment on “Global modelling of secondary organic aerosol in the troposphere: A sensitivity analysis” by K.Tsigaridis and M. Kanakidou***

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## **Major Comments**

This study offers results on sensitivities of global SOA production to factors such as the dependence of partition coefficients on temperatures and activity coefficients. However, the formulations for several of the sensitivity cases are either physically unrealizable or inconsistent with the chamber experiments of which the data were used in the study. The wrong formulations render parts of the manuscript irrelevant.

This study is correct in using Equation (20) instead of Equations (21) and (22) for the

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base case (simulation S1). The authors, however, are incorrect in their statement that Equation (20) underestimates SOA production. Starting from line 8 on page 2889, according to the manuscript, “this approach gives an upper limit for the evaporation process, since it assumes that all SOA evaporates from the particle at each timestep [sic].” While it is true that the mathematical solution to Equations (14), (25), and (20) is the same as the solution for the case if one assumes all SOA evaporates before re-partitioning, physically not all SOA necessarily evaporates. The term  $A_i^0$  in Equation (20) does not mean that the existing SOA has evaporated into the gas phase; it simply means that the  $A_i^0$ , in addition to  $G_i^0$  and  $\Delta G_i^p$ , is part of the condensible material, all of which need to be considered in the equilibrium calculation.

Equations (21) and (22), which are formulations for scenarios S2 and S4, are incorrect. The simplest way to see this is that if one combines Equations (21) and (22), one gets

$$\frac{A_i}{G_i} = \frac{A_i^0 + \frac{(G_i^0 + \Delta G_i^p)K_i M_0}{1 + K_i M_0}}{\frac{G_i^0 + \Delta G_i^p}{1 + K_i M_0}}$$

Clearly, the above does not satisfy the basic equilibrium condition (Equation (14)) unless  $A_i^0 = 0$ . Physically, Equation (21) is wrong because it assumes that once the semi-volatile products have partitioned into the aerosol phase, they become non-volatile. In reality, in the absence of chemical reactions in the condensed phase, they remain semi-volatile.

The scenario (simulation S1.3) in which the study considers “secondary” oxidation of the “primary” condensible products is inconsistent with the manner in which the chamber experiments in [3] and [4] were conducted. In the chamber experiments, oxidation has proceeded long enough that the parameters  $a_{ij}$  and  $K_{ij}$  correspond to final oxidation products, not first-generation products which the authors have assumed in this study.

It is not very clear from the wording of the manuscript, but it seems that this study

includes black carbon aerosol as part of the absorptive medium for SOA. This needs to be made clear. If the black carbon is pure graphitic carbon, this would not be the case as pure carbon cannot absorb organic material. In reality, the so-called “black carbon” does contain some organic materials that indeed can absorb SOA, but this has not been quantified. The authors should address this and maybe have a sensitivity case in which only primary organic aerosol is considered to absorb SOA.

In summary, cases S1.3, S2, and S4 should be removed from the paper, since these represent physically incorrect scenarios.

## Minor Comments

- On page 2883, line 20, the reference should be [2] instead of [1].
- In Equations (23) and (24), the authors should use a different notation as to not to confuse SOA partition coefficients with absorption rate coefficients.

## References

- [1] S. H. Chung and J. H. Seinfeld. Global distribution and climate forcing of carbonaceous aerosols. *J. Geophys. Res.*, 107(D19):4407, doi: 10.1029/ 2001JD001397, 2002.
- [2] W. F. Cooke, C. Lioussé, H. Cachier, and J. Feichter. Construction of a  $1^{\circ} \times 1^{\circ}$  fossil fuel emission data set for carbonaceous aerosol and implementation and radiative impact in the ECHAM4 model. *J. Geophys. Res.*, 104:22137–22162, 1999.

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- [3] R. J. Griffin, D. R. Cocker III, R. C. Flagan, and J. H. Seinfeld. Organic aerosol formation from the oxidation of biogenic hydrocarbons. *J. Geophys. Res.*, 104:3555–3567, 1999.
- [4] J. R. Odum, T. P. W. Jungkamp, R. J. Griffin, R. C. Flagan, and J. H. Seinfeld. The atmospheric aerosol-forming potential of whole gasoline vapor. *Science*, 276:96–99, 1997.

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