

## ***Interactive comment on “Naturally driven variability in the global secondary organic aerosol over a decade” by K. Tsigaridis et al.***

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Received and published: 13 March 2005

#### General comments:

The current paper deals with the annual variability of both anthropogenic and biogenic secondary organic aerosol (SOA) production influenced by meteorological parameters such as temperature and radiation. A special emphasis is put on simulating the biogenic precursor emissions by using the dynamic vegetation model ORCHIDEE to account for changes in and feedback processes of the biosphere. Moreover, the production of sulfate aerosols and the emissions of black carbon and primary organic aerosols have been included in the current study, whereas sea salt and dust particles have not been considered. With this set-up a period of 11 years including a year of

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spin-up time was simulated and the variability of SOA production and burden investigated.

In general, expected results such as increasing productivity with increasing emissions have been obtained. These have been presented and explained generally in a clear way with minor exceptions tackled below. Including the study of the SOA contribution to the global aerosol optical density this study reveals some key aspects of the important interaction of human-kind climate forcing and biological response and therefore is worth to be published in Atmospheric Chemistry and Physics.

#### Specific comments:

There are some details, which need to be explained in this paper to help the reader understand the simulated processes and to let him follow the individual explanations. These are:

a) A few words should be said about the chemical reactions and products taken into account. Although there are earlier publications cited for this purpose, this is essential with respect to the productivity of precursor compounds and with respect to the sinks of the formed aerosol particles. Is for example the SOA yield treated identical for all chemical conditions such as different  $\text{NO}_x$  and  $\text{HO}_x$  levels? Are individual species considered in the simulations and if so which water solubility do they possess? An identical contribution of different oxidation products to SOA mass production throughout the atmosphere independent on the chemical conditions would influence the water solubility notably, since for example in regions with high  $\text{NO}_x$  volume mixing ratios (e.g. North America, Europe, East Asia) the contribution of nitrogen containing compounds, with a low water solubility is higher, whereas in regions with high  $\text{HO}_x$  volume mixing ratios (at least sometimes in the tropics) the contribution of water-soluble oxidation products will lead to an increased water-solubility of the formed organic aerosol.

This applies as well for the statement about the interesting partial shift of SOA production away from the source region. What about anthropogenic SOA precursor com-

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pounds transported to a different chemical regime and the subsequent aerosol productivity?

b) What is the effect of the limitation of the dynamic vegetation model ORCHIDEE to ten different plant type functional groups? For sure, this model is a step forward compared to the approach using the GEIA datasets. But how does this influence the distribution of different monoterpene type leading to different rate constants with ozone, OH and  $\text{NO}_3$  and the productivity concerning aerosol production (compare e.g. limonene and  $\alpha$ -pinene) and the distribution of e.g. ozone [Bonn and Lawrence, 2005] ?

c) The particle size distribution is a quite important factor for calculating the dry as well as the wet deposition. Have this been approached by an average value of particle size or has this been treated by a modal approach?

Points a) and c) will influence the lifetime of SOA compounds remarkably, which is expected to vary between 1 and 10 days for  $\alpha$ -pinene oxidation compounds.

d) There is one interesting aspect, not of major importance but notably with respect to the simulation of sulfates for calculation the optical depth of atmospheric aerosols: In a recent paper, Spracklen *et al.* [2005] found that the inclusion of sea-salt, neglected in this study, was essential necessary to simulate sulphuric acid aerosol production, since it drives the cloud droplet chemistry of  $\text{SO}_2$ , and cloud condensation nuclei formation in the atmosphere. This might also affect SOA production by heterogeneous reactions, which nevertheless remains a topic of large uncertainty. What might be the impact of neglecting sea-salt in the present study, on the results obtained? Please discuss.

e) Finally there are two points, I did not understand in detail, most probably because of missing further information, according to the pints mentioned above. First, why is the rainfall reduced above the biogenic SOA source regions? Is this due to meteorological conditions, to the assumed low solubility of organic aerosol compounds or an increased number of particles such as black carbon containing aerosols caused by forest fires etc?

Second, when discussing the results obtained for the ratio Global SOAb burden to SOAb precursor VOC emission fro different years, I agree on the authors explanation

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for the lowest value determined for 1986 referring to the lowest temperature. But I am not convinced that the low values of 1990 and 1991 are actually caused mainly by the increased deposition flux (dry and wet), although it will contribute remarkably. By contrast, I would expect a major effect arising from the increased temperature and therefore lower productivity due to increased saturation vapour pressures of the oxidation products.

**References:**

Bonn, B. and Lawrence, M.G., Influence of biogenic secondary organic aerosol formation approaches on atmospheric chemistry. *J. Atmos. Chem.*, accepted.  
Spracklen, D.V., Pringle, K.J., Carslaw, K.S., Chipperfield, M.P., and Mann, G.W., A global off-line model of size-resolved aerosol microphysics: Model development and prediction of aerosol properties. *Atmos. Chem. Phys. Discuss.*, 5, 179-215, 2005.

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Interactive comment on Atmos. Chem. Phys. Discuss., 5, 1255, 2005.

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