

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

**K. Tsigaridis et al.**

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We would like to thank the referee for the time spent in reviewing the manuscript and for the thorough and useful suggestions on how to improve it. Here below is the point-by-point reply to the comments.

Detailed model description: The model has been described in great detail in an earlier ACP paper of our group (Tsigaridis and Kanakidou, 2003), available on the web. Therefore, we limited the explanations to absolutely necessary. A short paragraph has been added though at the end of section 2.1 to briefly mention the points noted by the referee: “The model parameterizations with regard to SOA production and fate in the atmosphere have been described in detail in Tsigaridis and Kanakidou (2003) and outlined below. The SOA concentration is calculated based on the equilibrium partitioning

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into an absorptive organic matter phase (Pankow, 1994a; 1994b; Odum et al., 1996). The partitioning coefficients of each SOA species are a function of air temperature and of the chemical composition of the aerosol on which it partitions. A two-product model approximation (Hoffmann et al., 1997; Odum et al., 1997) is being used to represent SOA formation on line with the VOC gas-phase chemistry. This approximation, contrary to explicit chemistry schemes (Derwent et al., 2003; Bonn and Lawrence, 2005), is the same both for high and low NO<sub>x</sub> environments and uses constant stoichiometric coefficients for the formation of semi-volatile products that lead to aerosols. This can be justified, since there is not enough understanding of the high NO<sub>x</sub> system with regard to SOA production (Kanakidou et al., 2005). The wet deposition of the gas-phase aerosol precursors depends on their solubility in cloud droplets. For the dry deposition of the gaseous species, the Ganzeveld and Lelieveld (1995) scheme has been used, which is based on the formulation developed by Wesely (1989). For the aerosol phase, wet and dry deposition is parameterized as suggested by Parungo et al. (1994) and already applied to sulphate particles in the model (Jeuken et al., 2001). Hydrophobic particles exhibit reduced dry deposition over wetted surfaces (Cooke et al., 1999) and are not removed in-cloud. All aerosols in the model have a rural continental size distribution, i.e. mostly accumulation range aerosol (Jeuken et al., 2001).”

Section 4 has been enriched by the following analysis of the effect of temperature, by adding the following in page 1265, first line: “Temperature changes affect almost all processes involved in the chemical production of SOA. They affect (i) the emissions (7% increase from 1986 to 1990), (ii) the reaction rates of the emitted VOC, thus affecting the rate of chemical production of the gas-phase aerosol precursors; this resulted in 0.3% enhancement of the contribution of O<sub>3</sub> reaction to the chemical loss of BVOC, (iii) the partitioning of organic matter between gas and aerosol phase leading to reduced production of the SOA with increasing temperature (minor reduction, <0.5%). The first two changes prevail over the last one due to the non-linear dependence of SOA production on the amount of preexisting aerosol mass.”

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The following text has been added in page 1265, line 20, to mention the effect of water absorption from hydrophilic particles (section 5): “It has to be noted that treating carbonaceous hydrophilic aerosols like sulfate, overestimates the amount of water associated to the aerosol, and has to be viewed as an upper limit of carbonaceous aerosol contribution to OD. The lower limit can be calculated considering dry aerosol. According our calculations, the aerosol associated water increases the OD by about 30%.”

The figures 6 and 7 have now been explained in more detail so the reader will get more information from them. For figure 6, the text (page 1265, line 23 until page 1266, line 3) has been replaced by: “The calculated OD of SOA (OD\_SOA) presents a high spatial (Figure 6) and temporal (not shown) variability, reflecting that of the emissions of biogenic VOC (7%), oxidant levels in the troposphere (5% OH, 4% O<sub>3</sub>, 4% NO<sub>3</sub> on a global scale) and meteorological conditions (mainly temperature and relative humidity) as discussed in section 4. The calculated differences in OD\_SOA resulting from changes both in meteorology and biogenic emissions (M90/E90 vs. M86/E86) are also shown in Figure 6 (lower panels) and reveal high variability with both positive and negative values and an overall effect of an increased global burden of SOA. The trend of increase or decrease of OD\_SOA follows mainly the trend of the emissions difference between 1986 and 1990. However, over some areas changes in rainfall that is the main aerosol removal process affect strongly the OD\_SOA (increased rainfall leads to reduced OD\_SOA).” While for figure 7, the text (page 1266, lines 4-7) has been modified as follows: “To further analyze SOA changes, we focus on O<sub>3</sub> that is the main oxidant involved in SOA formation in our model. The corresponding differences in the calculated tropospheric O<sub>3</sub> columns (from the surface to 300hPa) are shown in Figure 7. They reflect the increased production of O<sub>3</sub> in VOC limited areas when VOC emissions are higher due to higher temperature in 1990 than in 1986 and the enhanced loss of O<sub>3</sub> where relative humidity has been increased. In areas that were getting drier (Figure 1) O<sub>3</sub> has been increased, since the reaction of O<sub>1</sub>D with water vapor, acting as a net O<sub>3</sub> loss, has been depressed.”

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The last section of the paper is now called “Conclusions and need for future improvements”. A paragraph of future needed improvements has been added at the end of this section: “The work presented here has to be viewed as a first attempt to provide insight to the complexity of the climate/biosphere/atmosphere interactions involving SOA. The results largely rely on adopted parameterizations with regard to SOA formation. Improvements are needed to consider also the naturally emitted seasalt and dust particles. These particles could affect the chemical composition of the atmosphere by acting as surfaces for heterogeneous reactions that modify both the gas and aerosol phases. Multiphase chemistry and heterogeneous reactions on particles that produce particulate organic matter have to be considered in future modeling studies when appropriate experimental data will become available. Improved SOA formation parameterizations have to differentiate between high and low NO<sub>x</sub> environments (Bonn and Lawrence, 2005) and incorporate the hygroscopicity of the SOA components.”

The figures 2, 5 and 10 have been corrected, by using simple connector lines than smooth ones.

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