

Interactive comment on “Change in global aerosol composition since preindustrial times” by K. Tsigaridis et al.

K. Tsigaridis et al.

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We would like to thank Daven Henze and Robert Griffin for their comments, which motivate us to provide additional clarifications to the manuscript.

Indeed chamber experiments have shown that aromatics oxidation is highly dominated by the reaction of OH radicals. More than 99% of the oxidation of aromatic hydrocarbons proceeds via reaction with OH radicals in the TM3 model. Therefore the other pathways of aromatics oxidation turn to be negligible anthropogenic SOA sources. We will emphasize this in the final version, in order to avoid confusion.

SOA formation from biogenic VOC: It has to be noted that the oxidation of terpenes is governed by the O₃ reaction pathway, partly due to the increased reactivity applied in order to accurately represent the mixture of monoterpenes and higher homologues by

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only α - and β -pinene. This is described in detail in Tsigaridis and Kanakidou (2003). As a result, according to our model simulations, roughly 80% of the oxidation of monoterpenes occurs via the O₃ reaction. Further, the partitioning coefficients used in the present study are those reported by Presto et al. (2005) and not by Hoffmann et al. (1997) or Griffin et al. (1999) earlier works, as was the case in the Tsigaridis and Kanakidou (2003) paper. Presto et al. (2005) results have been used here in order to account for the change in SOA production when VOC/NO_x ratio varies around the globe. Presto et al. (2005) studied the ozonolysis of α -pinene, thus it wouldn't be appropriate to use the same SOA production efficiencies in the oxidation from other oxidants. Nonetheless, we made a sensitivity run assuming that the oxidation of α - and β -pinene from all three oxidants is able to produce SOA with the same efficiency as the ozonolysis reaction and found that the net chemical production of SOA has increased by 16% and the burden by 29%. We will include these numbers in the discussion of uncertainties.

Finally for clarity as asked by Henze and Griffin in the revised version of our manuscript, we will include additional explanations by further developing what is actually given in page 8, lines 16-24 and in Table 3, as briefly outlined hereafter.

The model always applies the NO_x-dependent yield parameters for the SOA formation from α -pinene and xylene oxidation. As mentioned in the manuscript, for β -pinene and toluene due to lack of NO_x-dependent data, the model applies modified aerosol yield parameters. These are calculated based on the independent SOA yield parameters (Griffin et al., 1999 for α - and β -pinene and Odum et al., 1997 for toluene and xylene) modified to provide SOA yields that present NO_x-dependence similar to those in the case of α -pinene and xylene, respectively.

The parameters taken from literature and used in the model are given in Table 3. In the revised version of the manuscript, for clarity, we will add the aerosol yield parameters for β -pinene and toluene “fitted” to the NO_x dependence of α -pinene and xylene respectively.

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The low and high VOC/NO_x regimes as defined from the chamber studies of Presto et al. (2005) for α -pinene and Song et al. (2005) for xylene are adopted in our study for biogenic and aromatic hydrocarbons, respectively. To characterize the regime under which SOA is formed, the model calculates at each time-step and for each of the SOA precursor hydrocarbons the VOC/NO_x ratio. Then it applies the appropriate aerosol yield parameters given in Table 3. This information that already exists in the manuscript but will be emphasized in the final version.

All above mentioned references are cited in the manuscript.

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