

## Response to Anonymous Referee #2

*This study describes the improvements the authors made for the simulation of secondary organic aerosols (SOA) using the updated SORGAM module, SORGAM-TIN. In the new module, the authors have successfully modified three parameters: the temperature dependence functions of SOA yields for aromatics and biogenic VOCs, OH initiated isoprene SOA, and isoprene and monoterpenes SOA formation channels from  $\text{NO}_3$  oxidation. The ambient SOA concentrations observed during the IMPACT field campaign in the Netherlands were used to evaluate the performance of SOEGAM-TIN. The updated model can reproduce the observed SOA much better than the original one, especially for those during nighttime. Possibilities of the daytime measurement-model discrepancies for the SORGAM-TIN model results were also discussed, although more discussion is needed. In general, the paper is written and organized clearly and concisely and is easy to follow. I recommend the acceptance of this paper for publication in ACP after some minor revisions shown below.*

**We would like to thank the reviewers for their comments and questions which have helped us to improve the manuscript. The reviewer's comments are given below together with our responses and changes made to the manuscript.**

*1. Page 5973, Lines 13-15: do you have any idea about the uncertainty raised from such an assumption? There should be significant differences among the spectra of the oxidation products of aromatics and biogenic VOCs with OH,  $\text{NO}_3$  and  $\text{O}_3$ .*

**Response:**

**Due to the differences of the oxidation products of the precursors with OH,  $\text{NO}_3$  and  $\text{O}_3$ , the extrapolation to other products may lead to uncertainties. For isoprene, the parameters for OH pathway and  $\text{NO}_3$  pathway were taken from published papers (Pye et al., 2010; Henze and Seinfeld, 2006) respectively, so it was different for the products oxidized by OH and  $\text{NO}_3$  radicals. For aromatics, OH radical was the main oxidant in both chamber experiments and atmosphere, so the uncertainty would not be large, especially as the reactions between anthropogenic precursors and  $\text{NO}_3$  radical were already included in the original model. Therefore, the key concern was the uncertainty for  $\alpha$ -pinene and limonene.**

**For  $\alpha$ -pinene and limonene, the simulated SOA in field study were both about 5% of the observation in the original model, and the main uncertainty was introduced with the products of  $\text{NO}_3$  pathways, according to the conditions of the experiments used in parameterization. In SORGAM-N, the simulated SOA by  $\alpha$ -pinene and limonene were**

about 17% of the observation, and the difference of the SORGAM-N and the original model was taken as the uncertainty caused by NO<sub>3</sub> chemical pathways for the two precursors. The uncertainty sits on top of the 24% discrepancy. Therefore, uncertainty of the extrapolation is smaller than 24%.

Please also see our answers to Specific comments #7 of referee #1.

2. Page 5974, Section 3.2: this section should be briefly mentioned in the abstract. In addition, it would be better to provide more information on the chamber experiment conditions such as the levels of biogenic VOCs, acidity and relative humidity, which may help understand the discrepancies among the chamber, model and ambient results discussed in the following section.

**Response:**

In chamber experiments for  $\alpha$ -pinene, the oxidant was ozone, and the concentrations of  $\alpha$ -pinene ranged from 45 to 692  $\mu\text{g}/\text{m}^3$  (average: 229  $\mu\text{g}/\text{m}^3$ ). In about half of the experiments, seed aerosol was used (as (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> or NH<sub>4</sub>HSO<sub>4</sub>). The largest relative humidity in the experiments was 73%, but in most experiments, the RH was lower than 50%, and in more than half of the experiments, the RH was very low (<10%).

For limonene, the main oxidant was ozone as well, and the concentrations of limonene ranged from 18 to 203  $\mu\text{g}/\text{m}^3$  (average: 112  $\mu\text{g}/\text{m}^3$ ). There were no seed aerosols in these experiments, and the RH was low than 10% in most of cases.

For m-xylene, the oxidant was OH radical, and the concentrations of m-xylene were in the range of 256 to 2114  $\mu\text{g}/\text{m}^3$  (average: 1123  $\mu\text{g}/\text{m}^3$ ). Sulfate seed aerosols were used in the experiments, and the RH was low (<10%) in more than 75% experiments.

We added these descriptions in the first paragraph in section 3.2.

3. Page 5978, Lines 4-7. The overestimations of SOA on May 9 and May 16 (Figure7a) seem to be largely from the failure of SORGAM-N. Is this true? Wet scavenging occurred on 16-17 May could strongly influence the atmospheric level of SOA.

**Response:**

According to the meteorological information and the simulations of other species, the reason of the overestimation of SOA on 16<sup>th</sup> of May should be the failure to simulate the wet scavenging. On 9<sup>th</sup> of May, SORGAM-N may be the main reason of the overestimation, with the highest value of NO<sub>3</sub> initialed SOA concentration simulated during the whole month. In this case, the extrapolation of the temperature dependent functions to NO<sub>3</sub> oxidation pathways should have larger error contribution indicating a need for further chamber

experiments on the NO<sub>3</sub> initiated SOA formations. It was revised as “However, the reason for the large discrepancy on 9<sup>th</sup> of May is probably due to errors of the SORGAM-N framework since the other parameters were well simulated in EURAD-IM system.” in L 9-11, P 5978.

4. Page 6005, in Figure 8a, SORGAM-TIN still strongly underestimates the observed SOA concentrations in the afternoon. The authors have discussed about some possible reasons in Section 3.3.3. However, special attention should be paid for isoprene derived SOA. The model result of the low level of SOA\_ISO in the afternoon as shown Figure 8b seems to be one of the main reasons for such an underestimation. Isoprene is known to be emitted during daytime and can be rapidly oxidized into semi-volatile organic species. Field campaigns have also shown that isoprene oxidation products such as 2-methyltetrols peaked during late afternoon to early evening due to a time lag of photo oxidation and gas/particle.

**Response:**

**We modified the discussions on this respect as the followings in page 5980 line 5:**

“... the parameterization of isoprene SOA formation process (e.g. the major SOA products were identified as 2-methyltetrols in both laboratory and field studies, cf. Claeys et al., 2004; Fu et al., 2010) could be possible reasons for the afternoon SOA underestimations with SORGAM-TIN as well.”

## **References:**

- Claeys, M.,Graham, B.,Vas, G.,Wang, W.,Vermeylen, R.,Pashynska, V.,Cafmeyer, J.,Guyon, P.,Andreae, M. O.,Artaxo, P. and Maenhaut, W.: Formation of secondary organic aerosols through photooxidation of isoprene, *Science*, 303(5661), 1173-1176, 2004.
- Fu, P. Q.,Kawamura, K.,Pavuluri, C. M.,Swaminathan, T. and Chen, J.: Molecular characterization of urban organic aerosol in tropical India: contributions of primary emissions and secondary photooxidation, *Atmospheric Chemistry and Physics*, 10(6), 2663-2689, 2010.
- Henze, D. K. and Seinfeld, J. H.: Global secondary organic aerosol from isoprene oxidation, *Geophysical Research Letters*, 33, L09812, doi:10.1029/2006gl025976, 2006.
- Pye, H. O. T.,Chan, A. W. H.,Barkley, M. P. and Seinfeld, J. H.: Global modeling of organic aerosol: the importance of reactive nitrogen (NO(x) and NO(3)), *Atmospheric Chemistry and Physics*, 10(22), 11261-11276, 2010.