Formation of secondary organic aerosol from isoprene oxidation over Europe

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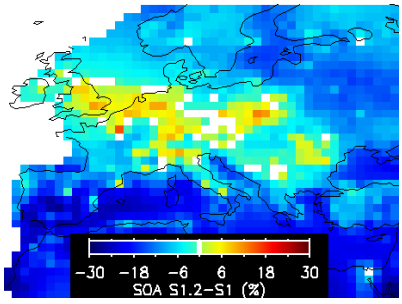
1. Introduction

The role of isoprene as a precursor to secondary organic aerosol (SOA) in the atmosphere over Europe was studied using the two-way nested global chemistry transport model TM5 [1] with a horizontal resolution of 1x1 degrees [2]. We analysed results from three scenarios: 1) reference scenario **S1**: similar to the study by Tsigaridis and Kanakidou (2003)[3] but including SOA formed from isoprene oxidation (SOA-I), 2) best guess scenario **S2**: considers several updates in parameterisations and uses the recent MEGAN isoprene emission inventory, and 3) zero SOA-I scenario **S3**: SOA formation from isoprene oxidation is ignored.

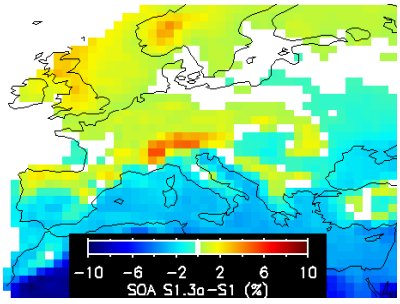
2. Sensitivity study

Two determining factors controlling SOA formation from isoprene in the gas-particle partitioning model have been selected for sensitivity analysis: the NO_x dependence of yields and partitioning coefficients and the temperature dependence of partitioning coefficients.

Applying the NO_x dependence of isoprene SOA yields leads to a decrease of SOA by 10-30% over most rural regions of Europe. However, the yield of the more volatile products from isoprene oxidation is roughly doubled under low VOC/NO_x compared to high VOC/NO_x. The increased concentration of gas phase products may lead to an increased SOA concentration, given that sufficient primary carbonaceous particles are available for condensation. This is the case over urban regions, in particular if these are downwind of forested areas.



The enthalpy of vaporisation, ΔH_{vap}, has been found to be a key parameter for the correct prediction of SOA concentrations, especially in the upper troposphere where temperatures are low [3, 4]. Using ΔH_{vap} = 72.7 $kJmol^{-1}$ for isoprene oxidation products instead of the standard value (42 $kJmol^{-1}$) leads to an increase of SOA over cold regions (Alps, UK, Norway) by up to 8%.



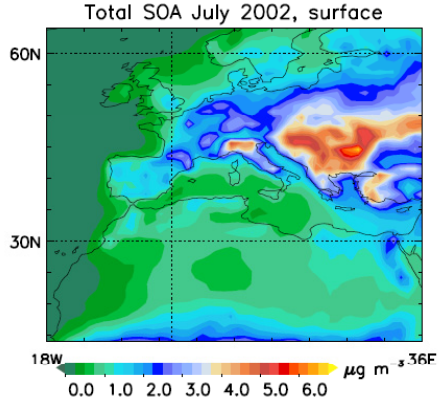
3. Results

The predicted tropospheric production of SOA-I over Europe using the best guess scenario **S2** is 0.10 Tg yr^{-1}. Total SOA production in this scenario is 0.70 Tg yr^{-1}, roughly 40% higher than in the zero SOA-I scenario (**S3**), highlighting the importance of secondary organic aerosol from isoprene oxidation in the atmosphere over Europe (see table below).

Between 7% and 60% of the SOA formed in the European PBL during summer is exported out of Europe, with 35% for **S1**. This corresponds to an export of 0.21 Tg yr^{-1} SOA out the European domain or - to a lesser extent - transport into higher regions of the atmosphere. Most exported SOA is advected from Europe to Asia. For comparison, Europe exports about 0.23 Tg yr^{-1} BC and 0.53 Tg yr^{-1} POC [5], with the majority of these aerosols being of anthropogenic origin. Thus the amount of SOA exported out of Europe is comparable to that of Black Carbon.

Table: Chemical production and burden of total SOA, isoprene derived SOA (SOA-I), SOA from terpenes (SOA-T), and anthropogenic SOA (SOA-A) in the European atmosphere (upper panel) and global atmosphere (lower panel).

Case	European production ($Tg yr^{-1}$)				European burden (Gg)			
	SOA Total	SOA-I	SOA-T	SOA-A	SOA Total	SOA-I	SOA-T	SOA-A
S1	0.98	0.40	0.51	0.07	48	23	22	3
S2	0.70	0.10	0.49	0.11	32	6	22	4
S3	0.51	0.00	0.44	0.07	24	0	21	3
Case	Global production ($Tg yr^{-1}$)				Global burden (Gg)			
	SOA Total	SOA-I	SOA-T	SOA-A	SOA Total	SOA-I	SOA-T	SOA-A
S1	32.9	15.2	16.9	0.8	230	119	106	5
S2	20.9	3.2	16.8	0.9	143	30	107	6
S3	16.9	0.0	16.1	0.8	114	0	109	5

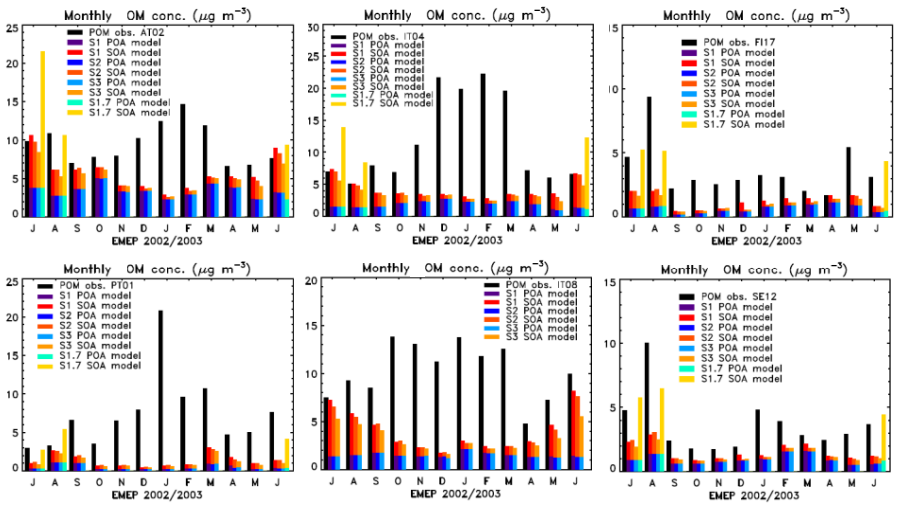


5. Conclusions

- Changes in the emissions of isoprene have a limited effect on SOA production rates over Europe (20-25%).
- Main fraction of organic aerosol observed at EMEP sites during summer is predicted to be secondary and of biogenic origin.
- Missing primary, wood-burning, organic particle sources in winter, are the most likely explanation for the wintertime discrepancy.
- Most urgent future research includes improvement of emission inventories (BC/POC), and experimental SOA studies on condensation behaviour, heterogeneous reactions and oligomerisation.

4. Comparison with measured OC from EMEP campaign 2002/2003

The best agreement of model and EMEP OC measurements [6] is found for Illmitz (AT02). While modelled particulate organic matter (OM) concentrations are in good agreement with the observations from April to October, a winter source of primary or secondary organic carbon particles is clearly missing. Similarly in Ispra (IT04) summer measurements are relatively well reproduced, while OM is strongly underestimated in winter. In Portugal (PT01), OM is underestimated throughout the year, but similarly to Ispra and Austria the strongest underestimation is in winter (by a factor of 20).



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