Atmos. Chem. Phys. Discuss., 5, S698–S701, 2005
www.atmos-chem-phys.org/acpd/5/S698/
European Geosciences Union

Atmospheric Chemistry and Physics Discussions

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

K. Tsigaridis et al.

Received and published: 6 May 2005

We thank Dr. Frank Dentener for his pertinent comments on the modeling work performed and the overall manuscript presentation. To address them, we have modified the text in the revised version as follows:

Abstract:

We agree with the comment. The sentence "Maximum values are calculated for 1990 (warmer and drier) and minimum values for 1986 (colder and wetter)." Has been added at line 15 of page 1256, together with the text "from 1986 to 1990" in the next sentence.

How much is the variability of SOA contributing to the variability of total aerosol:

The following text has been added at the end of the 2nd paragraph on section 3. "The

burden of all aerosols included in the model has increased by 1.8% from 1986 to 1990, with 0.8% attributed to the change in the meteorological conditions and the remaining 1% to the increase in SOAb burden (by 11.5%). Therefore, the natural variation of SOAb appears to be the most important contributor to the variation of the aerosol burden due to the natural changes. Note that seasalt and dust are not considered in the present study."

What is the consistency between the meteorological fields used in the TM3 model and those in ORCHIDEE:

In page 1261, line 15 the following discussion has been added: "For the present study ORCHIDEE has been forced by ISLSCP2 satellite observations (http://islscp2.sesda.com/ISLCP2_1/html_pages) suitable for use in models of the biosphere. Note that the ECMWF assimilated ERA-15 dataset has been used by the TM3 model. These datasets (ISLCP2 and ECMWF) are both based on observations, limiting therefore possible inconsistencies between the derived biogenic emissions and the TM3 simulations."

It would be good to have a paragraph describing the difference of ORCHIDEE; with the well know Guenther et al. fields.

In order to clarify differences between the Guenther et al. (1995) and ORCHIDEE fields, in page 1260 before equation 1 the lines 3-5 the text has been modified as follows: "Biogenic emissions of VOC are calculated based on the well-known Guenther et al. (1995) parameterizations and take into account additional features such as the impact of leaf age on isoprene and methanol emissions. On top of the isoprene, monoterpenes, and other volatile organic compounds (OVOC) emissions considered in previous works, ORCHIDEE also calculates explicitly biogenic emissions of methanol, acetone, acetaldehyde, formaldehyde, acetic and formic acids." Additionally, in page 1261 lines 5-7, the text has been modified: "Since the ORCHIDEE model does not calculate the leaf temperature, we use instead the "surface" temperature, that reflects

ACPD

5, S698–S701, 2005

the radiative budget including soil surface and canopy, and not the air temperature, as done by Guenther et al. (1995). The GEIA inventory for isoprene and terpenes is 20%-30% higher than that of ORCHIDEE. This difference is below the uncertainty of the factor of 2 to 3 associated with the BVOC emissions."

page 1257 line 15: This is a more general statement for atmospheric composition change. The discussion on page 1257 lines 8-16 have been rephrased as follows, to refer explicitly to SOA: "A critical step in understanding the behavior of trace constituents in the atmosphere is the evaluation of the importance of the human induced changes that presupposes the understanding and evaluation of the natural variability (see discussion on CO2 in Falkowski et al., 2000). In particular, for SOA although the anthropogenic emissions enhance both ozone and preexisting aerosols (Tsigaridis and Kanakidou, 2003), the natural variability of biogenic VOC emissions could affect SOA levels as much as the anthropogenic emissions. The variability of SOA levels in the atmosphere exerts in turn, together with the other aerosol components, an impact on radiation and on the hydrological cycle (Kanakidou et al., 2005 and references therein). Thus, SOA might be involved in significant chemistry/climate feedbacks that enhance stability or perturbation of the atmosphere and climate, and that deserve careful investigation (Kulmala et al., 2004)." This part has been moved in line 8 of page 1257.

There are several studies focusing on O3 and OH variability in TM3 connected to ERA15. Peters et al. [JGR, 2001] and Dentener et al. [JGR, 2003]:

This could indeed be a very good point of discussion, but it is not the topic of this paper. Our first results are consistent, although slightly different from these earlier studies with the TM3 model.

Difference in boreal Russia:

In figure 3, top right panel the big positive difference in the emissions from 1986 to 1990 reflects the corresponding difference in temperature for the same period. Temperature is the critical parameter responsible for the emission differences, as shown in equation

ACPD
5, S698–S701, 2005

3. This is explained at the end of the first paragraph in section 3: “The increase in the emissions from 1986 to 1990 reaches 7% in all VOC categories, and is a result of the natural variability of the climate system (i.e. temperature and light intensity which are key parameters in biogenic emissions variability, and water cycle) with generally colder and drier 1986 than 1990 (Figure 1). These temperature and water cycle differences are not uniformly distributed around the globe; therefore the calculated emission changes vary spatially (Figure 3)”.

Anthropogenic emissions and ozone boundary conditions:

The following sentence has been added in page 1258, line 22 (section 2.1): “All anthropogenic emissions (reference year 1990) were kept constant from year to year, while ozone boundary conditions vary, based on TOMS data.”

Optical parameters for SOA and SOA interaction with RH:

The extinction efficiency for all aerosols in the model are shown in Table 2 (as mentioned in Section 5) and the SOA interaction with RH is based on a polynomial fit by Veefkind (1999), again mentioned in Section 5.

Finally, in agreement with Frank Dentener’s last comment we added the following sentence to the conclusions: “Therefore, based on current knowledge, the meteorological factors determining the SOA production cause variability far below the uncertainty of the SOA formation itself (more than a factor of two; Tsigaridis and Kanakidou, 2003).”

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