

Interactive comment on “Organic aerosol and global climate modelling: a review” by M. Kanakidou et al.

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We would like to thank Dr. Alex Guenther for his constructive and pertinent comments on the manuscript. We have incorporated all his suggestions as follows:

A short section on aerosol lifetimes has been added in page 5898: Lifetime of carbonaceous aerosols The lifetime of carbonaceous aerosols driven by wet and dry deposition depends on the affinity for water of the aerosols (whether they are hydrophobic or hydrophilic, see discussion in section 5). The main sink process for these aerosols is wet deposition which represents 70 to 85% of the total sink for these species; the other remaining sink is dry deposition. The AEROCOM intercomparison (<http://nansen.ipsl.jussieu.fr/AEROCOM/> global aerosol model intercomparison) that includes the results of 16 global models which represent the main aerosol com-

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ponents showed a range of lifetimes of organic aerosol from 4.3 to 11.1 days with 13 out of these 16 models lying between 5.4 and 8.1 days. The main cause for the variations of aerosol lifetime is the spatial (latitude and altitude) and temporal variation of the precipitation and the vicinity of surfaces for dry deposition. Integrated monthly, the latitudinal variations in lifetime can be as large as a factor of 4 (Balkanski et al., 1993). The variations with altitude are of similar importance since for an aerosol produced in the first kilometer of the troposphere the globally mean lifetime is of approximately 4 days whereas when the aerosol is formed at 10 km its lifetime increases to 12 days (Balkanski, 1991)

Balkanski Y., D. Jacob, G. Gardner, W. Graustein, and K. K. Turekian. Tropospheric Residence Times of Continental Aerosols derived from a Three Dimensional Simulation of 210Pb, *J. Geoph. Res.*, 98, pp. 20573-20586, 1993. Balkanski, Y.. Atmospheric residence time of continental aerosols, Ph.D. Thesis, Harvard University, 1991.

Unfortunately, constraining the global SOA production remains quite challenging. Most current estimates are based on measurements of the OC concentrations, the fraction of the OC that is primary and the lifetime of OC in the atmosphere. All these (as described in various sections of the paper) are quite uncertain. There are relatively few measurements with significant uncertainties due to sampling and analytical issues. The fraction of the organic aerosol that is secondary is quite uncertain especially in the more polluted continental regions. Finally the lifetime of the OC is a lot more uncertain compared to that of the inorganic salts. The result is that these constraints based on observations result in estimates of the global SOA production are as uncertain as these of the bottom-up estimates. This discussion has been added at the end of a new section 12.5 on the SOA measurements (comment by A. Gelencsér,). This demonstrates the large uncertainties and difficulties in evaluating the SOA modeling.

The following statement (as suggested by A. Guenther) is now added as is in page 5864, line 16 “ Note however that recent studies report that $> C_7$ carbonyls may be important contributors to SOA (Matsunaga et al., 2003). Current analytical methods

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may need to be improved before we can accurately quantify these compounds.”

The Griffin et al (1999) rough estimates are based on compound specific emissions from each plant, major plant species identification per ecosystem and global BVOC emission estimates by ecosystems based on the Guenther et al (1995) work. To make this clearer to the reader we rephrased the sentence in page 5865 as follows: “ Seinfeld and Pankow (2003) combined the Griffin et al. (1999b) ORVOC breakdown in smaller chemical categories and classify the SOA forming compounds in a total of 11 categories, which encompass on a global scale most of the observed terpenes and ORVOCs emissions, with the Guenther et al. (1995) mass emissions.” We also added the word ‘about’ in line 16 page 5864 to strengthen the uncertain character of this percentage. No change is made in Table 1 caption where both Griffin et al. and Guenther et al. references are cited.

A paragraph has been added at the end of the section 2.2.4 page 5868: “Anthropogenic VOC emissions are 5-10 times lower than biogenic VOC emissions (excluding CH₄). When considering the known SOA precursor emissions alone, this ratio increases above 10. Large uncertainties exist also in the anthropogenic emission factors for SOA precursors like aromatics and some oxygenated solvents. Global inventories of NMVOC anthropogenic emissions are generally calculated in two steps. First, using an emission factor approach, total NMVOC emissions are calculated on the basis of (inter)national activity statistics (e.g. fuel consumption, solvent use) and emission factors that take into account abatement technologies. According to Olivier et al. (1999b), the uncertainty in total NMVOC emissions has been estimated to be ~50% for fossil fuel related emissions and ~100% for non fossil fuel emissions. Second, the total NMVOC emissions are generally subdivided in specific NMVOC species clusters. Toluene, xylene and trimethylbenzene are three different NMVOC groups for which an emission profile has been defined in the EDGAR database (Oliver et al. 1996). For each NMVOC profile an activity specific profile has been defined, which assigns the fraction of each of the NMVOC group to the total NMVOC emis-

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sions. In general global uniform NMVOC profiles are based on data from USA and EU countries (e.g. <http://www.epa.gov/ttn/chief/software/speciate/index.html> or http://reports.eea.eu.int/EMEP_CORINAIR3/en). Application of these global aggregated NMVOC profiles leads to another considerable uncertainty. Specific quantitative uncertainty estimates on toluene, xylene and trimethylbenzene are -to our best knowledge- not available at the moment.”

We decided not to convert the BVOC emissions to SOA by a constant fraction to avoid a misinterpretation of this information. Since the SOA is formed in the whole troposphere the application of a constant factor to the boundary layer emissions of BVOC over simplifies reality. The secondary source of SOA is extended to the whole troposphere and well above the boundary layer (BL). This is crucial for SOA fate since the lifetime of aerosol is larger in the free troposphere than in the boundary layer as discussed in section 4.3 (of the revised version). The vertical distribution of the SOA source is already discussed a few lines further in the same paragraph and at the end we have added the last sentence pointing to the importance of this fact.

When using the 100% SOA yield hypothesis in page 5866, line 18, we have now added the comment: ” (higher than reported by Griffin et al., 1999b),”

Since there is no information available to construct a table as suggested by A. Guenther, the following sentences have been also added in page 5874, line 17: - There is very little and fragmentary information available about the SOA yields from sesquiterpenes. These yields are much higher than those of the monoterpenes (17-67% on a mass basis reported by Griffin et al., 1999b) and based on these yields the contribution of sesquiterpenes to SOA global formation may be up to 9% (Griffin et al. 1999a).

The following comment has been added in section 5.1 to address the validation of dry deposition process. “The evaluation of dry deposition removal of aerosols in the models remains quite uncertain since it would require eddy-correlation measurements of carbonaceous aerosols which to our knowledge do not exist at present.“

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The sentence has been modified to “Most important components in the group ‘terpenes’, after isoprene that is a hemi-terpene, are the mono-terpenes (C₁₀H₁₆) α-pinene, β-pinene, sabinene, and limonene (Table 1), accounting for 40-80% of the overall terpene emission on a global scale when isoprene is excluded.”

MEGAN work is now mentioned and the corresponding web site is provided.

Page 5869, the sentence has been changed to ‘invoke integration of Ě’

Page 5887, ‘Quarantees’ has been replaced by ‘provides’. Other small suggested corrections/additions have been also performed.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 5855, 2004.

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