

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

M. Kanakidou et al.

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We would like to thank referee #6 for his useful comments to the manuscript. To address them we have performed the following changes/additions:

Two sentences have been added in page 5865 lines 23 and 25. The text now reads: From sector analysis it follows that traffic, industrial processes and solvent use were responsible for large decrease. In Europe, reported emissions decreased in the EU from 16 kT y⁻¹ in 1989 to 12 kT y⁻¹ in 2000 (EMEP, 2003). Latter trends are confirmed by a limited number of measurements (Monks et al., 2003). Reductions have been reported for acetylene, ethane, benzene and toluene (Roemer, 2001).

The comment for Table 2 in page 5866 lines 2-5 has been rephrased: “Table 2 gives

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the regional break-down of the primary anthropogenic organic aerosol (POA) flux estimates by Bond (2004) and terpene and ORVOC emissions. Note that the importance of oceans (O'Dowd et al., 2004) and of vegetation as natural sources of POA remains to be determined.”

Further in page 5866, line 26 the sentence has been modified to “Ė small fraction (about 10%) of the global SOA formation Ė, although locally might be much more important. This contribution is of the same order of magnitude with the naturally driven variation of the SOA chemical production (Tsigaridis et al., 2005).” Tsigaridis, K., Lathiere, J., Kanakidou, M., Hauglustaine, D. A.: Naturally driven variability in the global secondary organic aerosol over a decade, in press ACPD-2004-0285, 2005.

In page 5869 line 13 the following sentence has been added: “In particular the potential important POA marine source from the ocean currently omitted from climate -modelling prediction should be evaluated (O'Dowd et al., 2004)”

Two sentences have been added at the end of 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.”

Title of section 2.3 has been changed as suggested to “Uncertainties and Research Needs”.

Section 2.3. The word ‘natural’ has been added for clarity.

Section 3, introduction: The chain (processes leading to SOA formation) has been complemented by both aqueous phase chemistry and cloud processing.

Section 3.1.1 This is a good comment. At the end of the paragraph, we added the following text “The ability of most SOA compounds to nucleate in the atmosphere is not well understood and is the topic of current research. One needs to understand not only the volatility of these compounds but also their surface energy because of

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the importance of the Kelvin effect for nucleation. Note, that the increase in molecular weight leads the greater Kelvin effect (Seinfeld and Pandis, 1998) that hinders the homogeneous nucleation. Therefore, for an organic species to homogeneously nucleate, the lowering of vapour pressure when increasing functionality with increased molecular weight should overcome the counteracting Kelvin effect.”

The following text has been added in section 3.2: “For the anthropogenic emissions the processes leading to nucleation and SOA will probably be much more varied than for the biogenics because of the greater diversity in the chemical structures of the compounds emitted. There are experiments (Johnson et al., 2005) which support that ozone is not a major player in the aromatic systems and that nitroaromatic compounds may be playing a dominate role. For anthropogenic species containing double bonds, however, the evidence here again (Tobias et al., 2000; Kalberer et al., 2000) supports that ozone reactions are probably the most important for SOA formation.”

Johnson, D., Jenkin, M. E., Wirtz, K., Martin-Reviejo, M.: Simulating the Formation of Secondary Organic Aerosol from the Photooxidation of Aromatic Hydrocarbons, *Environ. Chem.*, in press, 2005. Kalberer, M., Yu, J., Cocker, D. R., Flagan, R. C., and Seinfeld, J. H.: Aerosol formation in the cyclohexene-ozone system, *Environ. Sci. Technol.* 34, 4894-4901, 2000. Tobias, H. J., Docherty, K. S., Beving, D. E., Ziemann, P. J.: Effect of Relative Humidity on the Chemical Composition of Secondary Organic Aerosol Formed from Reactions of 1-Tetradecene and O₃, *Environ. Sci. Technol.*, 34, 2116-2125, 2000.

Section 3.2, we added the following comment in the text. “A potential explanation for this could be that most low-vapor-pressure organics have high molecular weight, but high-MW species have a strong Kelvin effect at nucleation sizes as discussed in section 3.1.1.”

Section 4 is moved inside section 3 before section 3.3

Explanation of “organic phase” has been added in the beginning of the section.

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Section 4 is now before section 3.3

There is no doubt that the Jacobson JGR 2004 paper is very interesting and the work done is state of the art, however it does not treat SOA formation as SOA is traditionally defined (chemically produced in the atmosphere). It only considers the primary OM and a reversible inorganic chemistry. The condensation of organic gases that have been emitted as primary aerosols, which evaporated, is different from the SOA chemical formation. Therefore it is not included in Table 5.

In page 5897, line 1 for clarity the word 'mass' is added: "Ěby which organic aerosol mass is scavengedĚ"

Notwithstanding the description of wet deposition in global model an equivalent scavenging efficiency can be derived for both the in-cloud and below-cloud process. What we wanted to bring the attention of the reader to is that in the case of snow flakes, the efficiency of collection of the aerosol is widely varying and only a model that predicts the shape of the snow flakes can account for this important effect. We added this last comment in page 5898, line 17 together with the ref to the Jacobson JGR 2003 paper mentioned by the referee.

The sentence in section 6.1. has been changed to: "Presently, most models Ě"

"some hours after emission" has been added in the last sentence of section 6.2

It is correct that eq. 18 is only valid for a single particle. We have modified the sentence preceding eq. 18 from 'The single scattering albedo is defined as:' to 'The single scattering albedo of a single particle is defined as:'

It is correct that the complex index of refraction is a property of a compound. However, in a particle containing more than one chemical constituent, the complex refractive index depends, among other, upon the chemical composition, that is the concentration of the individual components. The referee mistakes particle concentration and chemical component concentrations in the particles.

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We have slightly modified the sentence by removing reference to 'concentration': "Only a single study exists on the complex index of refraction of various organic aerosol components and mixtures representative of atmospheric conditions."

Reference to Jacobson, (2003, 2004) is added as small correction suggested by point 14 of the referees comments.

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

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