

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

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This article presents a thorough up-to-date review and discusses crucial gaps in our knowledge on secondary organic aerosol formation from terpenes (including isoprene). The information presented in this review will be useful as a basis for future interdisciplinary research efforts. Following are some comments/suggestions that the authors may want to consider in revising their article:

Section 2.1 - page 5862. You write: “There are a number of other types of primary carbonaceous material in the atmosphere such as viruses, bacteria, and plant debris.” I suggest to add “fungal spores” which are known to be airborne and to be present in the PM10 fraction. As a reference you could use Bauer et al., 2002.

Section 2.2.1 - page 5863. You write: “Very recent studies, however, indicate involvement of isoprene as source for SOA (Jang et al., 2003a; Claeys et al., 2004a). I suggest to include at this stage the following two studies:

- Limbeck et al., 2003 (cited elsewhere in the paper): this laboratory study clearly demonstrates that isoprene is a precursor for SOA in the presence of sulfuric acid, more specifically, for humic-like substances. I anticipate that humic-like substances originating from isoprene oxidation contribute more to the SOA mass than the 2 methyl-tretols which we reported recently (Claeys et al., 2004a);

- Matsunaga et al., 2003 (mentioned in the interactive comment by A. Guenther): this field study carried out at a Japanese mixed forest site shows that the semi-volatile isoprene oxidation products, glycolaldehyde and hydroxyacetone, were important SOA constituents.

Section 3.1 - Chemistry of secondary aerosol formation. Edney et al., 2003 showed that PM_{2.5} aerosols collected at a semi-rural mixed forest site in the southeastern USA contains significant amounts of a C₈ tricarboxylic acid, which could be generated in smog chamber experiments of an alpha-pinene/air mixture with NO_x but not with ozone. These results suggest that SOA from irradiated alpha-pinene/NO_x/air mixtures better represents the PM_{2.5} aerosol at this semi-rural forest site than that generated by ozonolysis of alpha-pinene. This is explained by hydroxyl radical initiated reactions in the irradiated mixture, and/or the likelihood that the irradiated system included more secondary reactions leading to the highly oxidized SOA. The alpha-pinene oxidation product, the C₈ tricarboxylic acid, reported by Edney et al., 2003, has first been chemically characterized by Kubátová et al., 2002, in urban aerosols from Gent, Belgium, as well as in rain forest aerosols from Amazonia, Brazil. In our experience, there is clearly a discrepancy between the nature of alpha-pinene oxidation products formed in chamber experiments with ozone and those found in field studies.

You write on page 5875 - line 11: “Until recently dicarboxylic acids were the lowest

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volatile compounds positively identified in terpenes generated aerosol.” I suggest to update this with data on the formation of the C8 tricarboxylic acid which is more polar than dicarboxylic acids; this could perhaps be done on page 5876, after your statement “There is, however, evidence that compounds less volatile than dicarboxylic acids are present in the aerosol.”

Section 3.1.4 - Concluding remarks. Here, I suggest to add:

- of the impact of SO₂ levels on the SOA formed through heterogenous reactions: photo-oxidation of SO₂ results in sulfuric acid which is a catalyst in the formation of humic-like substances from isoprene and alpha-pinene (Limbeck et al., 2003) as well as in the formation of 2-methyltetrols from isoprene (Claeys et al., 2004b).

Section 14 - Conclusions. I would not write that ozonolysis reactions are the most important for SOA formation. Based on our own results showing that 2-methyltetrols are significant polar SOA products at a rural continental mixed forest site, i.e., K-pusztá, Hungary (Claeys et al, 2004b), it appears that other reactions such as acid-catalyzed oxidation reactions with hydrogen peroxide in the aqueous aerosol phase may be important for SOA formation. Furthermore, based on the results obtained by Edney et al., 2003 (discussed above) and our own findings on alpha-pinene oxidation products detected in PM_{2.5} forest aerosols from different sites (Amazonia, Brazil, and K-pusztá, Hungary), it appears that oxidation reactions of alpha-pinene induced by NO_x may be as important as those induced by ozone for explaining alpha-pinene oxidation products found in ambient aerosol.

References:

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