Interactive comment on “Secondary organic aerosol in the global aerosol – chemistry transport model Oslo CTM2” by C. R. Hoyle et al.

Anonymous Referee #1

Received and published: 18 July 2007

This paper describes the implementation of a secondary organic aerosol (SOA) simulation into a global model (Oslo CTM2). The authors include SOA production from monoterpenes, ORVOCs, isoprene and aromatics following previous global model implementations, particularly Chung and Seinfeld (2002). The global budgets are summarized for a baseline simulation and a simulation where SOA condensation takes place on ammonium sulfate as well as organic aerosol. Simulated surface concentrations are also compared to a subset of global observations. This manuscript describes a thorough implementation of SOA in a global model and the authors attempt to explore interesting elements of the simulation, in particular the importance of NO3 oxidation to SOA formation. The paper is clearly written. However, the manuscript often fails to adequately explore the results obtained. As a result, this study, which is heavily based
on previous global model studies appears limited in originality. Discussion of previous work is also occasionally lacking or limited. Comments included below should be addressed before publication in ACP.

Specific Comments

Pg 9055, In 29: Tsigaridis and Kanikidou 2003 included the same anthropogenic aromatic precursors considered in this study, not “predominantly natural precursors”

Pg 9056, ln24-25: How does this study reduce the uncertainty in global SOA production and burden? What new sensitivities will be explored here? How does this work go beyond Chung and Seinfeld, Henze and Seinfeld and Tsigaridis and Kanakidou?

Pg 9058: The decision to treat monoterpene emissions with the same temperature and light dependence of isoprene deserves further discussion here. The continuing emission of monoterpenes at night has primarily been measured in North America, whereas recent studies with European species (as cited by the author) have found that monoterpenes may respond similarly to isoprene (ie. light sensitive). I believe that it is generally thought that this discrepancy lies in plant speciation, but that this is unconfirmed. As this study represents the first time (as far as I’m aware) of a global model study implementing light-sensitive monoterpene emissions, it would be valuable to provide further details on how this will affect the simulation. In particular, perhaps the distribution of all monoterpene emissions into the daylight period effectively enhances (doubles?) the daytime SOA formation from previous studies where emission was diurnally constant with light.

Pg. 9059: Why do the authors treat the light sensitivity as a function of solar elevation angle, rather than as a function of photosynthetically active radiation (PAR) as in Guenther et al.?

Pg. 9059, ln. 18-19: Why did the authors add the emissions of other aromatics into the emission estimates of toluene and xylene? Is this based on evidence that trimethyl-
benzene, and all aromatics form SOA? From what studies?

Section 2.4: What ratio of organic matter to organic carbon was used here?

Section 2.5: What molecular weights for the SOA formed were used here?

Table 4: Where are these rate constants from? In particular, the rate constant for toluene+OH seems fast compared to Atkinson and Arey (2003).

Table 5: Include the citations for the values used here.

Table 7: How is the Henry’s Law coefficient for aromatics estimated? Or what reference used?

Section 3: The authors should acknowledge (and compare to) the previous work of Tsigaridis and Kanakidou (2003) who explored the enhanced SOA production when ammonium sulfate and nitrate were used as condensation mass for SOA.

Section 3.1: SOA production from monoterpenes and ORVOC alone is 3 times higher than in Chung and Seinfeld (2002) with the exact same mechanism and similar precursor emissions. This requires further investigation. Unlike the burden, SOA production is not affected by the removal mechanisms in these different models. The authors should perform a sensitivity study with the same 1.15 day time-scale for aging to verify the importance of this effect. In addition, I recommend that the authors qualitatively compare their POA distributions to those published by Chung and Seinfeld (2002) to identify any obvious differences.

Section 3.1: Discuss the simulation performed where all SOA partitions to aerosol. What are the global simulated SOA production and burden? How high were POA emissions to force this complete conversion to aerosol?

Pg. 9065, In 4-5: The condensation of SOA on ammonium sulfate appears to produce a proportionately greater increase in SOA from anthropogenic precursors than biogenic. Presumably this is due to the coincident emission locations of these anthropogenic
compounds? Discussion of this (or other reasons) should be included here.

Pg. 9065, In 22: Explain why the lifetime of SOA increases in the 2nd model simulation.

Pg. 9066, In 13: “latitude and longitude” and altitude?

Pg. 9067: Further to the author’s discussion on representativeness - What kind of sites are these? Several of the sites, particularly in the third set, appear urban and may be impossible to represent with a global model.

Pg. 9068, In 7: The underestimate of SOA may also be the result of missing SOA formation mechanisms.

Pg. 9068, In 10-11: If the Oslo CTM2 simulation of sulfate is to be used as validation of the wet deposition algorithms the authors need to include either figures or numbers showing this comparison with observations or cite a study which shows this comparison.

Section 4: The NO3 results appear counter-intuitive (short-lived terpene emissions are only emitted during daytime in this simulation and NO3 is only significantly produced at night - so how is SOA formed?) and require further discussion. In particular, why is this higher than in previous studies (as stated in the conclusions, In 10)?

Pg. 9070, In 22: Laboratory results have not provided guidance as to whether SOA formation on ammonium sulfate follows the same absorption as in organic matter, therefore it is inappropriate to assume that including this effect in the model is an “improvement”. Of course SOA formation is enhanced with this effect, improving agreement with observations, but it is not possible to ascertain whether this is an accurate treatment.