Interactive comment on “Development of a secondary organic aerosol formation mechanism: comparison with smog chamber experiments and atmospheric measurements” by L. E. Olcese et al.

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This paper is dealing with the modeling of secondary organic aerosol formation in the troposphere that is a scientific topic that attracts a lot of attention the recent years. Therefore it is relevant to ACP and would deserve publication only after addressing a number of points that will clarify the applied procedure, support and justify the results. In addition to the detailed comments of the other referee, I would like to strengthen the need for the following clarifications/justifications:

1. Why do the authors adopt the non evaporative approach? The fact that it fits better the results is not a justification. Actual knowledge shows that oligomerisation and heterogeneous reactions can push deviating from the gas to particle equilibrium of the
pure SOA component by lowering the vapour pressures of the aerosol phase SOA components. Adopting a non evaporative approach does not account for molecular mass changes that might be related to those processes and should be only viewed as a sensitivity test and not as a base case simulation. // True, here we essentially assume that the molecular weight does not change in the organic aerosol phase due to polymerization. We have added a discussion and results about the two approaches. A sensitivity study shows that changes of about 50% in the MW do change the final SOA concentration in less than 3%. We have added a discussion to the paper.

2. As a consequence, the authors have to also shown the results with the equilibrium approach and plot them in all figures of the paper together with the non-evaporative approach results. At the end it might turn out that the model is not performing as well as mentioned in the paper if a more rigorous comparison procedure is followed. // We have added the results and plots for evaporative and non-evaporative schemes.

3. In addition, the authors claim (page 8373, line 3) that their results show reasonable agreement with observations without any adjustment to the partitioning coefficients derived from theory. If a non evaporative approach is used - this is a major adjustment! // In our simulations this is not a major adjustment for the total SOA because the partition coefficients are so large. There is no evidence to favor one scheme or the other, and the non-evaporative scheme takes account in a crude way of possible reactions within the aerosol phase to produce less volatile species.

4. Another area of large uncertainties in the calculations is the removal rate of the aerosols (page 8374, line 5). The authors provide an aerosol removal time constant of 7 days and a boundary layer height of 5000 m. How is the estimate of 7 days derived? For a global mean removal rate that in the case of aerosols is mainly due to wet removal processes this number is reasonable but what about the specific experiment? Can the authors provide information on the specific conditions that govern the area in March 1st? In addition the boundary layer looks huge, might be a typo? Mixing in such a high continental boundary layer (constant day and night?)
could minimize the effect of dry deposition. More details and thorough discussion are needed at this point. // We reported incorrectly the mixing height of 5000 m, the real value is 1500 m. The estimation for the 7 day residence time in the atmosphere is taken from the Kanakidou et al. (2005) work and AEROCOM intercomparison results (http://nansen.ipsl.jussieu.fr/AEROCOM/, 2006), and are referenced in the text. Since the specific meteorological conditions for these simulations are not available, we added a sensitivity test using a removal time appropriate to only dry deposition (35 days) (now described in the paper), but since the simulations are only for 3 days, total SOA increased only by 2%. The ambient simulations do not intend to predict the total organic aerosol concentrations found in particular urban-influenced or rural sites, but are performed only to have a rough estimation of SOA production under those conditions.

5. page 8374, line13: The difference of 10% between the continuous equilibrium assumption and the non evaporative approach looks too small. Again reporting both simulations in the figures would provide a better view to the reader of the importance of this assumption. // The referee is right about the need for an explanation for the different schemes used. We added this explanation, as well as the results of the calculations and a plot including the results of the evaporative and non-evaporative schemes.

6. page 8374, line 23: Here the authors state that biogenics account for about 90% of the total SOA mass whereas in the conclusion this number is approximately 60% (page 8377, line 22). // The 60% is the percentage obtained in the TORCH simulations, whereas the 90% value was for the urban-influenced and rural simulations. We had a mistake in one reaction in the chemical mechanism and in one partitioning species that leaded to wrong values in some of the simulations; the right percentage of biogenics in the urban-influenced simulations is 67%. We have clarified the differencet between biogenics percentage in the text.

7. page 8378, line 6 to 15: this is a rather general statement and definitely not a conclusion, should be earlier in the introduction or the main core of the paper. // The referee is right, paragraphs moved to the ambient simulations section.
8. page 8378, lines 16-21: What about a remote marine site? What about Mexico City? A few more case studies could support the validity of the modeling approach. // The SOA production at a remote marine site is very low (about 6 x 10^{-4} \text{?g m}^{-3} \text{ in a three days simulation}), so we did not think this was appropriate to add to the study. Mexico City is an interesting case but beyond our scope.

9. page 8378, line 22-25: How do the authors extrapolate for remote site cases? This part is not rigorous, concrete procedure and numbers have to be provided or the sentence has to be removed as very speculative. // We agree that this part is not extremely rigorous, but works as a simple approximation. The SOA production in remote sites is very low, so it is not included; all the recent work focuses the SOA production in urban areas (anthropogenic sources) and in rural/forest sites (biogenic sources). We modified the text to better explain what we did.

10. page 8362, line 9: what is here stated as an 'average error' is mainly the deviation of model results from the observations. // Yes, we will modify the text to make it clearer. Page 8362, line 9, add after ...an average error of 16%... : "..., compared with the experimental results."

11. page 8378, line 27: Tsigaridis et al (Atmos. Chem. Phys., 6, 5143-5162, 2006) considering that all oxidants can contribute to SOA formation with the ozonolysis reported yields have estimated only a 16% increase in the global annual biogenic SOA leading to a 29% increase to the corresponding burden. On the other hand, Hoyle et al (open discussion paper, Atmos. Chem. Phys. Discuss., 7, 9053-9092, 2007) estimate that NO3 reactions account for 50%-60% of the total amount of SOA near the surface in polluted regions and less than 25% in more remote areas. In both studies the global estimates are far below the 235 Tg/y given as upper limit in the present study. // The mistake in the chemical mechanism gave a high global SOA estimation, now the results are still higher than other estimations (63 Tg/y), but lower than our previous estimation. The main difference between those estimations and ours is the role of the isoprene. Recent studies (Henze and Seinfeld, GRL 2006) have shown that the global SOA pro-
duction from isoprene is higher than that predicted from the two products scheme that was used in the Tsigaridis et al. paper. From the Henze and Seinfeld paper "Inclusion of isoprene as a source of secondary organic aerosol (SOA) in a global model increases the global burden of SOA from all sources by more than a factor of two."

12. Table 1, title of the 2nd column: specify what 'significant' means, provide numbers. // The referee is right, header changed and explanation added in Table 1.

13. Table 2, provide ozone levels also. // Add, at the end of line 2 in page 8371: "The initial ozone concentration was equal to zero in all the 28 simulations."

14. page 8363, line 15: and different SOA precursor molecules speciation. // The referee is right, phrase added.