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Atmospheric
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Interactive comment on “Modeling secondary organic aerosol formation through cloud processing of organic compounds” *by* J. Chen et al.

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The paper presents an integrated approach to evaluate the importance of cloud organic chemistry in SOA formation. Despite its limitations, the model confirms that the previous assumptions that were based on the observations of individual compounds or on very schematic model calculations are indeed valid and organic reactions occurring in clouds do contribute to SOA on a regional and possibly global scale. For the first time, some quantitative estimates are also provided for the more complex three-dimensional simulation. There are, however, some concerns regarding the structure of the model, in particular of the zero-dimensional one. There are large inhomogeneity in the level of complexity of the model: while gas-phase reactions are treated in much details, and part of the aqueous-phase mechanisms up to carbon number four

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are also based on exact kinetic models, a very large fraction of potential organics with carbon number greater than four (which together could be more important in SOA formation than the few low MW compounds) are treated as surrogates using very simple assumptions (Page 8956 line 10-11). Basically, it is assumed that aldehydes are converted into their corresponding carboxylic acids in clouds. So it is to some extent trivial that cloud reactions that decrease the volatility of a great many compounds in surrogates will inevitably increase the amount of SOA formed. Larger MW aldehydes (e.g. pinonaldehyde) are generally assumed to contribute directly (but only in a small fraction due to their volatility) to SOA mass, and in OH reactions they do not oxidize further but even decompose to small gaseous species. Thus cloud reactions which not only preserve these abundant species but convert them into less volatile (and more stable) species, will add mass to SOA, no question about it. Thus the entire complex model is pending on a single and simple assumption which still awaits confirmation in chemistry. This would make the model an order of magnitude estimate at best, but certainly would not justify reporting percentage increase up to 3 significant digits (such as Page 8964 Line 20). In their paper, the authors do not expressly emphasize this important limitation and its consequences on the entire model study. They simply refer to it as part of the modeling approach, and give reference to a relevant but not very recent paper (Aumont et al., 2000). Furthermore, among the model assumptions, clouds seem not to affect gas-phase photolysis at all (Page 8959, Line 11-12). In addition, clouds are also treated in a very simplistic way: monodisperse clouds are assumed to occupy the entire boundary layer of 1000 m height for three hours each day. It would have been interesting to see how important cloud reactions remain if clouds occupy part of the boundary layer and affect actinic flux as they normally do. In general, the paper is very valuable as a comprehensive modeling study, but its limitations must be explicitly stated (also in the Abstract and Discussion) and the conclusions need to be formulated in the light of these limitations.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 8951, 2007.