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Interactive comment on “Development of a simple unified volatility-based scheme (SUVS) for secondary organic aerosol formation using genetic algorithms” by A. G. Xia et al.

Anonymous Referee #3

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The authors present a new approach to reduce the organic gas phase oxidation schemes to describe the time evolution of the volatility of secondary organic species produced from the α -pinene oxidation. The approach is fully empirical and uses the MCM chemicals scheme as a reference scheme. A conceptual chemical scheme is first designed (the simple unified volatility based scheme - SUVS). The yields of the various products and the rate constants in the SUVS are next tuned in order to minimize the difference of the simulated volatility distribution. A genetic algorithm method is used to optimize a set of more than 400 parameters included in the SUVS. The paper show that the SUVS reproduce well the volatility distribution simulated with the MCM for various scenarios. The approach used to reduce the scheme is original and clearly

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presented. However, the paper raises many issues that should be addressed in the revised version of the paper to be published in ACP.

How is the inorganic gas-phase chemistry handled? The SUVS scheme must likely be plugged to another chemical scheme to describe the time evolution of the HOx/NOx/Ox system. From the SUVS scheme described in fig. 1, this information is required to compute the lifetime of the organic species (XHC, S) as well as the branching ratio of the peroxy surrogate species (XO₂). It would be helpful to include in the paper few sentences explaining how the SUVS is connected to the gas phase chemical scheme (if any).

Does the SUVS modify the Ox/NOx/HOx budget? I understand that SUVS is only designed for the purpose of SOA modeling. However, the scheme described in Fig. 1 suggests that the SUVS scheme modify the HOx/NOx/Ox budget (e.g. part 2 appears as a NOx loss, reaction 35 as a HOx loss, ...). If that is the case, then the authors should discuss the reliability of the reduced scheme with respect to the key inorganic species (e.g. O₃, OH, NOx, ...). On the other hand, if the SUVS scheme does not modify the budget of the inorganic species, then how is the HOx/NOx/Ox budget handled in the simulation including the SUVS? The α -pinene oxidation in the reference MCM scheme not only leads to a set of secondary organic products but also drive the NOx/HOx/Ox budget.

How reliable and useful is the SUVS scheme? The reliability of the SUVS scheme is directly linked to the accuracy of the MCM scheme. The MCM includes the current knowledge of the gas phase oxidation of the organic compounds. However, most reactions accounted in the MCM are based on structure/activity relationships and subject of significant uncertainties. In addition, the MCM species are sorted in the various bins according to estimated vapor pressure. The reliability of the final SUVS also depends on the accuracy of the method used to estimate these properties. Furthermore and as pointed by the authors, the species of interest are produce after many oxidation steps. The chemistry of most species produced after few oxidation steps is described in a

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simplified way in the MCM. I don't think that the MCM (or any other detailed/explicit scheme) can be considered as a faithful representation of the actual atmospheric processes leading to SOA formation. It would therefore be useful that the paper includes a short discussion that put in perspective the reliability of the final SUVS.

Is the SUVS approach sustainable? The SUVS is here optimized for the α -pinene oxidation at 298 K. The optimization process must be run whenever the MCM scheme is updated. The optimization process seems to be too expensive to be performed systematically. I have the feeling that, at least for exploratory simulations, this computational cost could be used to directly solve the MCM in a 3D models (see for example Jacobson and Ginnebaugh, JGR, 2010, DOI:10.1029/2009JD013289 or Ying and Li, Atmos. Env., 2011, doi: 10.1016/j.atmosenv.2011.03.043). Furthermore, the authors claim that "the same SUVS with update parameters could be used to describe the SOA formation from different precursors". Does it mean that:

- for each parent compound taken into account, a specific SUVS (i.e. 31 species) is added to the chemical scheme? The computational cost to provide the set of parameters for the various parent compounds seems prohibitive.
- a single SUVS can be tailored for various mixtures of primary species? In that case, is there good reason to believe that a reliable single set of parameters can be used to describe various mixtures of parent compounds?

Can we learn something from the optimization process? As described by the authors, the design of the SUVS is based on the key gas phase oxidation steps. Can the optimized parameters be interpreted? Are they within the range of the "expected" values? It would be useful to publish the values of the parameters as supplementary material added to the paper.

Can some properties be assigned to the set of lumped condensable species? The SUVS is here assessed on a molar basis. A final user will likely more be interested in SOA mass concentration. Can a molar mass be assigned to the 10 lumped condens-

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able species? For later applications and comparisons with observations, the O/C and N/C ratio provide very useful constrain. Can a typical oxygen and nitrogen content be assigned to the SUVS?

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 3885, 2011.

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