

This paper develops 2p-VBS modeling parameters, which are based on VBS framework, but are computationally efficient compared to the original VBS from which it was derived.

First I do not see any major advantages to the development of a 2p-VBS scheme, without consideration of “further” multi-generational chemistry. This is because the original VBS parameterizations [Donahue *et al.*, 2006; Robinson *et al.*, 2007] were developed to include “further” multi-generational chemistry of organic vapors. Any development of a computationally efficient scheme or with a “reduced” parameter VBS could only be acceptable after it has been tested with the original VBS including “further” multi-generational chemistry. A good example of this development was presented by Shrivastava *et al.* [Shrivastava *et al.*, 2011]. Shrivastava *et al.* [2011] evaluated their reduced 2-parameter VBS species (1-parameter for traditional biogenic and anthropogenic species) against predictions from the 9-species VBS including multi-generational chemistry. Second, the authors do not discuss or even acknowledge the most recent papers and developments in SOA field specifically related to the low volatility, high viscosity and the semi-solid nature of the SOA particles in several recent studies [Abramson *et al.*, 2013; Cappa and Wilson, 2011; Perraud *et al.*, 2012; Shrivastava *et al.*, 2013; Vaden *et al.*, 2011; Vaden *et al.*, 2010; Virtanen *et al.*, 2010; Zelenyuk *et al.*, 2012]. It is important to at least comment on the implications of their 2p-VBS parameters if SOA was semi-solid. Using the 3D chemical transport model WRF-Chem, Shrivastava *et al.* [2013] showed that there could be large differences between the semi-volatile liquid-like and semi-solid SOA modeling paradigms in the atmosphere, for parameterizations including multi-generational chemistry with fragmentation. In addition, their box model showed that these differences could be large even for non-aging parameterizations especially under cleaner conditions. The authors here presented CMAQ simulations (3D chemical transport) without even acknowledging the previous studies on semi-solid SOA behavior.

Due to these major shortcomings I do not find this paper acceptable in the present form, and recommend major revisions, accounting for comparisons including multi-generational chemistry of SOA precursors.

Besides these major issues, there are several other issues which need to be fixed:

1. Introduction: Lines 15-20: How can brown carbon lead to negative radiative forcing? Brown carbon is supposed to be absorbing and should show a warming effect. This has to be clarified
2. Page 15913: last paragraph: The authors included  $H_{vap}$  as an additional fitting parameter following Shrivastava *et al.* (2008). This statement is misleading because Shrivastava *et al.* (2008) did not fit  $\Delta H_{vap}$ , rather they considered  $\Delta H_{vap}$  varying with the volatility bins similar to Donahue *et al.* (2006).
3. Table 2: I disagree with calling POA in the work of Shrivastava *et al.* 2008 as “undefined POA”. In their preceding work, Shrivastava *et al.* [2006] showed that partitioning behavior of both diesel and wood smoke could be described by similar parameters. Since these two very different sources could be described by similar partitioning parameters, they applied the same set of VBS parameters to all POA sources in Shrivastava *et al.* 2008.

4. Page 15914: Paragraph 15: The comparison of 2p-VBS with the reduced 2-species VBS in Shrivastava et al. (2011) should be removed both from the main text and the supporting information. This is not a meaningful comparison for 2 reasons: (a) Shrivastava et al. (2011) used the 2-species VBS only for the non-traditional SOA precursors which are subject to multi-generational aging (SVOC and IVOC precursors from sources including fossil and biomass burning). The authors here do not include further multi-generational chemistry (b) Shrivastava et al. (2011) used only 1-species for the traditional biogenic precursors and found their predictions to be low. However, this was not very critical in their study as biogenic SOA was less important compared to the other sources for their MILAGRO 2006 case study, also discussed in other studies (e.g. Hodzic et al. [2010]). Thus this comparison is confusing and does not add anything meaningful to this study
5. Section 3.3. CMAQ model simulations: The authors should include “further” multigenerational chemistry of both 2p-VBS parameters and the original VBS to show relative differences.
6. Figure 9: This figure is not central to the paper and could be a part of Supplemental Information.
7. Table 3: It is important to also include the specific measurements from which the best available parameters were derived, as footnotes.

## References

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