The authors sincerely thank the reviewers for their thoughtful and thorough reviews. Responses to the *reviewer comments* (in italics) appear below.

Reviewer #3

General: *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" multigenerational 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.*

In this study we focus on SOA formation that occurs on timescales that have been parameterized based on chamber studies (~ < 10 hours); our work, and others, clearly show that improvements in the representation of SOA formation at these timescales are needed. We have made detailed comparisons of widely-used 2p and VBS parameterizations with one another and with published chamber data, focusing on the low M_o range that is relevant to ambient conditions. Our work demonstrates that limitations in current parameterizations have implications for results obtained from 3D models and improvements can be made. Comparisons between model-based secondary organic carbon (SOC) and measurement-based SOC show that using the "best available" parameters (low NOx) reduces the percent bias from -63% to -15% (please see response to Reviewer #1, comment #1).

We agree with the reviewer that multi-generational chemical processes cannot be ignored in parameterizations of SOA/POA formation. However, we think that one can and should separate the multi-generational chemistry that occurs on timescales that can be parameterized based on chamber studies (~ < 10 hours), and the multi-generational chemistry that occurs on timescales that have not yet been captured in such studies (~ < 3 days). Most of the discussion in our manuscript focuses on the formation of "traditional" SOA, often called V-SOA (e.g., Shrivastava et al., 2011), from the precursors listed in Table 3. We note here that as in our work, Shrivastava et al. (2011) did not treat the aging of gas-phase V-SOA species in the WRF-Chem/VBS model because of the reported findings of Dzepina et al. (2011) that doing so resulted in significant overprediction of SOA on a regional scale; they also noted the uncertainty of developing aging parameterizations based on chamber data since such timescales are not accessible in chambers. A recent publication by Henry et al. (2012) discussed SOA formation in two stages, stage 1:"first generation" in which oxidation products are lumped (via 2p or VBS approach) and SOA mass yields are parameterized for individual VOCs or lumped VOCs (based on emissions categories); and stage 2: "aging" in which first generation oxidation products are further oxidized in the gas phase by OH (aging also can include photolysis and accretion reactions). Henry et al. (2012) stated: "…it is important to constrain the first-generation chemistry for precursor VOCs driven by each oxidant" and "As yields are currently parameterized in CTMs, we must try to isolate the first generation chemistry as much as possible for the initial formation of SOA". While the Henry et al. (2012) paper largely discussed the need for chamber experiments that facilitate parameterization of these processes separately, from a modeling perspective we too advocate the need to accurately define the "starting point" for aging parameterizations.

In this work, we did not modify the treatment of aging within the CMAQ model. CMAQ currently treats all POA as nonvolatile (no "SI-SOA" formation) and semi-volatile SOA is converted to non-volatile SOA with a rate constant of 9.6×10^{-6} s^{-1}. We recognize that this treatment of aging is different than that in most implementations of the VBS, including the reduced species VBS described by Shrivastava et al. (2011). We did carefully analyze widely used parameterizations (2p and VBS) of "first-generation" SOA formation based on chamber experiments and devised a list of "best available" parameters. Recommendations were made to replace some of the default CMAQ parameters with the newly developed 2p-VBS parameters; SOA predictions using the "best available" parameters in CMAQ were evaluated. Implementation of the 2p-VBS parameters in a CTM employing the VBS approach, not done in this work, would of course require evaluation.

In response to the reviewer we have modified the manuscript to better describe the difference between the 2p-VBS parameters developed here and the reduced VBS parameters in Shrivastava et al. (2011). Revised in the introduction: "The 2p-VBS parameters derived in this work are fundamentally different from the reduced species VBS parameters of Shrivastava et al. (2011), **in which a single volatility bin ($C^* = 1$ μg m^{-3}) is used to represent SOA formation from the traditional precursors**, also developed to reduce computational burden specifically in CTMs running online meteorology. Differences between the approaches and resulting parameters are further described in the supplementary material (see Figure S2)." Added to Section 2.2: "The best available parameters represent "first generation" SOA formation, i.e., SOA formation that can be parameterized based on chamber experiments (see Henry et al., 2012). Aging/processing in CMAQ is treated by converting the semi-volatile SOA (formed by the traditional precursors) into non-volatile SOA with a rate constant of 9.6×10^{-6} s^{-1} (Carlton et al., 2010)."

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 multigenerational 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.]

The work of Shrivastava et al. (2013) will be important for shaping the treatment of condensed-phase diffusion limitations to instantaneous equilibrium partitioning in CTMs. While we do not support adaptation of the Shrivastava et al. (2013) approach in all CTMs at this time, we recognize our oversight in not mentioning these important publications. Please see the response to Reviewer #1, comment # 3.

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.

To our knowledge, detailed investigations of: 1) widely used SOA parameters, including as affected by the fitting approach (2p vs. VBS) and data source(s); and 2) the ability of those parameters to represent chamber data more broadly (i.e., including data not used for fitting parameters) have not been published. Our study has highlighted the limitation of current parameterizations in representing yields of "traditional" SOA at atmospherically relevant M_o loadings. While multi-generational chemistry is important, it is beyond the scope of our focus on traditional SOA.

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*

   We agree this sentence was poorly worded and confusing. For the revision, please see the response to Reviewer #2, major/minor comment #1.

2. *Page 15913: last paragraph: The authors included Hvap as an additional fitting parameter following Shrivastava et al. (2008). This statement is misleading because Shrivastava et al. (2008) did not fit deltaHvap, rather they considered deltaHvap varying with the volatility bins similar to Donahue et al. (2006).*

   We have rewritten the sentence to more accurately describe the work of Shrivastava et al. (2008): "In Shrivastava et al. (2008), ΔH_{vap} for POA varied with C^*; here we introduced ΔH_{vap} as an additional fitting parameter so that the temperature-dependence of the full VBS yield curves would be represented by the 2p-VBS yield curves. For the 2p-VBS, ΔH_{vap} values are included in Table 2."

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.*

   We have replaced "undefined" with "undifferentiated" to more accurately describe the use of a single set of parameters to describe diesel and wood smoke POA.

4. *Page 15914: Paragragh 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 multigenerational 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

In agreement with this reviewer, we believe it important to distinguish between the “2p-VBS” parameters in this work, and the “2 species VBS” parameters introduced by Shrivastava et al. (2011). We have revised the text of the supplement to focus only on the traditional SOA precursors, which can be directly compared between the two studies. We agree with the reviewer that the discussion of the POA *C** values may be confusing to readers and is not directly applicable. Shrivastava et al. (2011) provided 1 volatility bin parameterizations (also called 1-species, 1-product), $C^* = 1\ \mu g\ m^{-3}$, for both biogenic and anthropogenic precursors (not just biogenic as the reviewer suggests). These parameterizations are based on chamber studies, and not subject to any further aging, and thus should be directly comparable to the 2p-VBS parameters developed in this work. Such comparisons are particularly relevant in this work, since it is the linkages between the chamber experiments, the parameters, and the CTM output that we are interested in; that the Shrivastava et al. (2011) parameters did not impact that particular modeling study significantly is less relevant to our work.

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.*

   As noted previously (and now clarified in the manuscript), CMAQ, and other CTMs implementing the 2p framework, do not treat aging the way that it is treated in CTMs employing the VBS framework. The default aging scheme of CMAQ was retained in this work. We also reiterate that it is not VBS parameters we are suggesting to replace, but 2p parameters. Please see also first response under “General”.

6. *Figure 9: This figure is not central to the paper and could be a part of Supplemental Information.*

   We disagree with the reviewer on this point. Given the significantly higher SOA mass predicted for the low-NOx case, it is important to consider where in the ambient atmosphere such low-NOx conditions may be relevant. To the best of our knowledge, such a figure has not been published.

7. *Table 3: It is important to also include the specific measurements from which the best available parameters were derived, as footnotes.*

   To the best of our ability, we have included data sources, either in the text and/or figures. Please also note the compilation of experimental data in Table S1. The best available parameters are based on published parameterizations, as indicated by the last column in

Table 3; the data can be found in he individual references, Carlton et al. (2010) and Tsimpidi et al. (2010), now listed in Table 3.

References:

Henry et al., *Environmental Science & Technology,* 46: 12347-12354, 2012