Referee 1:

Terms IVOC, LVOC, SVOC, etc. are used multiple times (e.g. p. 17299, Fig. 1) but are not defined here (definitions are provided in Donahue et al., 2012). I suggest providing a brief definition/description of these terms.

We have added a description in the caption of Figure 1.

Figure captions, especially figures 2-6, tend more towards discussion than description. I would suggest adding a sentence at the beginning of each to describe what is being plotted.

We have added additional descriptions to Figures 2-6.

Specific Comments:

p. 17292 Eq. 2: Do nc and no correspond to the number of carbons and oxygens, respectively? ***Yes. We have added an explanation of these terms after the equation.***
p. 17298 lines 6-7: Is there atmospheric relevance to values of 1 and 0.15, e.g. typical rural and urban values? ***NO_x concentrations in remote areas can be as little as 2-8 ppt, while concentrations in urban areas range from 10-1000 ppb (Seinfeld and Pandis, 2006). This creates a range of β from 1 (where essentially none of the RO_2 radicals are removed by NO) to 0 (where essentially all of the RO_2 radicals are removed by NO), respectively.***
Fig 2: I assume the blue lines for OM are for high-NOx conditions and would suggest indicating this on the legend. ***We have added a sentence clarifying this in the caption of the figure.***
Fig 3: Suggest noting in the caption the order of magnitude difference in the COM scales between plots (a) and (b). ***We added a sentence in the caption noting the order of magnitude difference.***

Technical Corrections:

p. 17285 line 4: etc. Organics ***Corrected***
p. 17285 line 25: organic compounds ***Corrected***
p. 17286 line 11: NO 3 has not [been] defined ***We have added the definition in the previous paragraph.***
p. 17286 lines 17-18: most commonly ***Corrected***
p. 17287 line 27: 30:46 can ***Corrected***
p. 17291 line 4: MATLAB (throughout) ***Corrected***
p. 17296 line 9: phase (Trump and Donahue, 2014), ***Corrected***
p. 17296 line 15: left to a later ***Corrected***
p. 17297 line 22: A concentration of 100 μg m - 3 ***Corrected***

Referee 2:

Page 17289, Line 22: I think the VBS approach needs more than 4-7 species to be transported in 3-D air quality models. The number of advected species in 3D models depends on the implementation of the VBS approach, nevertheless at least 10-20 species are needed to be transported for implementation of the VBS approach in air quality models.

A species here denotes a representative species for a single volatility bin. A basis set that uses 6 volatility bins would have 6 surrogate species. Depending on the implementation of the VBS, a 3-D model may use multiple instances of a basis set. Each instance can represent a different aerosol class or source. For instance, Koo et al. (2014) implements 5 species in its basis set each for SV-OOA, HOA, and BBOA. Using the 2D-VBS, the modeling of organics can be written as a single accumulation of mass regardless of source, or it can be treated separately.

Equation 7: How much POA do you assume in your calculations?

The model simulates a chamber experiment of α-pinene aging and does not contain POA.

In your calculations OH levels do not change depending on the NOx levels, right? Doesnt this introduce inconsistencies when the model is applied to ambient conditions? The same is true for other species, the 2D VBS model doesnt take into account chemical reactions between gaseous species. One of the ultimate goals of such detailed box modeling studies is to improve the SOA parametrization in air quality and climate models. My main comment is how one can use the findings of this updated 2D VBS box model in air quality models for SOA? I suggest authors to add estimates for sensitivity of SOA yields to NOx levels for major SOA precursors, e.g. toluene, long chain alkanes, terpenes etc. How the branching ratio calculation introduced by Lane et al. (2008) can be improved by using the results of this study? Since the full implementation of the 2D VBS approach in 3D models is not feasible due to the very high number of species that need to tracked, the authors should provide some guidelines how such complex and detailed SOA models could be applied in more simplified SOA parametrizations, which are currently used in a number of air quality and climate models.

A 3-D model is not the only case where the VBS is useful. This model is concerned with simulating the specific chemistry of α-pinene aging that is studied in chamber experiments. During these experiments, the levels of reactants are held constant to understand the sophisticated chemistry that organic precursors undergo to form SOA. Thus, we are not making any statements about ambient concentrations of NO_x, and the exploration of NO_x on other organics is beyond the scope of this work. The next step in modeling more computationally intensive chemistry is Lagrangian plume tracking. This was first

implemented in PMCAMx by Murphy et al. (2012) to model the EUCAARI campaigns. It has also been used in CMAQ by Koo et al. (2014) and Zhao et al. (2015).

References

Koo, B., Knipping, E., and Yarwood, G.: 1.5-Dimensional volatility basis set approach for modeling organic aerosol in CAMx and CMAQ, Atmos. Environ., 95, 158–164, doi:10.1016/j.atmosenv.2014.06.031, 2014.

Murphy, B. N., Donahue, N. M., Fountoukis, C., Dall'Osto, M., O'Dowd, C., Kiendler-Scharr, A., and Pandis, S. N.: Functionalization and fragmentation during ambient organic aerosol aging: application of the 2-D volatility basis set to field studies, Atmos. Chem. Phys., 12, 10 797–10 816, doi:10.5194/acp-12-10797-2012, 2012.

Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, Second Edition, John Wiley & Sons, Inc., 2006.

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