Response to Interactive comment by Anonymous Reviewer 2

Original comments in **black**. Reponses in **blue** and proposed new text is *italicized*.

The manuscript "Simulating secondary organic aerosol in a regional air quality model using the statistical oxidation model – Part 2: Assessing the influence of vapor wall losses" by Cappa et al presents a study of how wall-losses of secondary organic aerosol (SOA) in chamber measurements affect the modeled atmospheric SOA concentrations. There have been several studies on wall losses in chamber measurements. However, to my knowledge this is the first study that has comprehensively taken chamber wall losses into account in a large scale atmospheric model. The manuscript fits well in the scope of Atmospheric Chemistry and Physics and very well written. I can recommend it to be published after the following minor issues have been addressed:

We thank the reviewer for the comments. Our responses to specific queries follow below.

1. Abstract, Page 30083, Line 28, "Similar improvements...": This sentence is difficult to understand without reading the whole manuscript. In addition, in Section 2.3.2 it is said that with more volatility bins, "wall-less" fits could be determined. Although the results of this study strongly indicate that wall-losses have to be taken into account in order to reproduce observed SOA properties, I would recommend excluding this sentence from the Abstract.

We agree that this sentence may be difficult to understand without the context of the entire manuscript, and will delete this sentence in the abstract upon revision.

2. Page 30089, Line 25-: Doesn't the accommodation coefficient have any effect on the amount of SOA or is the effect insignificant?

The accommodation coefficient can influence the simulated amount of SOA. However, if the original parameterization was developed using the same accommodation coefficient as is used in the 3D model, then much of the effect will inherently be accounted for. Consider that in (Zhang et al., 2014) we found it was possible to fit laboratory data for individual experiments with good fidelity using a range of accommodation coefficients by altering the other model parameters to account for the variation in the net accommodation rate. If, however, one were to develop a parameterization using a particular value of the accommodation coefficient but then were to perform simulations using a different value then certainly the simulated SOA concentration would be dramatically influenced. In the current work, we have developed the parameterization using the same accommodation coefficient as is used in the 3D simulations, and thus we do not expect that the accommodation coefficient would directly influence the amount of SOA simulated in the model. There is, however, an indirect effect in that an assumption of smaller accommodation coefficients during the parameterization development (i.e. during data fitting) leads to a larger influence of vapor wall losses (McVay et al., 2014; Zhang et al., 2014). As noted on P30090, Line 5 (and discussed further in our response to Dr. Pye), we assume that the gas-particle accommodation coefficient is unity here, and thus these simulations represent a conservative estimate (likely lower estimate for a given assumed k_{wall}) of the influence of vapor wall losses on the simulated absolute SOA concentrations. A similar question was raised by Reviewer #1, and we refer the reader to our Response to Point 3 to Reviewer #1 for additional discussion.

3. Page 30091, Line 11: This inconsistent behaviour should be explained. Now it is only shown in Fig S2 and not really explained anywhere.

We will elaborate on this idea. More specifically, by "inconsistent behavior" we mean that some fits were "good," some were "okay" and some were quite obviously "poor." These are of course qualitative statements, although do convey the point shown in Fig. S2. We will modify the specific sentence to read:

> Thus, when fits were performed, inconsistent behavior between the different vapor wall loss conditions was obtained over the atmospherically relevant concentration range (~ 0.1–20 μg m^{-3}) *in that some of the fits matched the data well over the entire range while other fits deviated strongly from the observations, especially at lower SOA concentrations.*

4. Page 30093, Line 29: How is SOA formation from isoprene a notable exception?

By "notable exception" we mean that the HO_2/NO dependence of the SOA yields has been explored in detail for this system. To clarify our point, we will modify the sentence to read:

> (SOA formation from isoprene is a notable exception *in that the dependence of SOA formation on HO_2 and NO has been examined in detail* (e.g. Xu et al., 2014).)

5. Page 30095, Line 26: Should this be along the lines of "R_{wall} increases with decreasing SOA concentration"?

Yes, we can modify the sentence as suggested to make it clearer.

6. Page 30096, Lines 4-7: Is this true for both relative and absolute differences? I would expect that the absolute differences in concentrations between the wallloss and no-wall-loss simulations are higher in high-source regions.

We agree with the reviewer that the absolute differences are likely to be larger where the absolute concentrations are larger. Our focus is more on the relative difference, or more specifically the relative model/measurement difference. We will modify the sentence to read:

> Additionally, it has been suggested that the typical underprediction of SOA by air quality and chemical transport models *relative to observations* might increase with photochemical age (Volkamer et al., 2006). The current results suggest the possibility that the SOA concentrations in more remote (lower concentration) regions may be underestimated in models to a greater extent *in a relative sense* than in high-source (higher concentration) regions due to a lack of accounting for vapor wall losses, *although the absolute differences in SOA concentrations may be larger in regions where absolute concentrations are larger*.

7. Page 30102, Line 1: It is unclear to me, what is the "fossil fraction of SOA".

The fossil fraction of SOA is the fraction of the total SOA that is sourced from fossil-derived VOCs (e.g. toluene, alkanes) as opposed to biogenic VOCs (e.g. isoprene, monoterpenes, sesquiterpenes). We will state this more explicitly as:

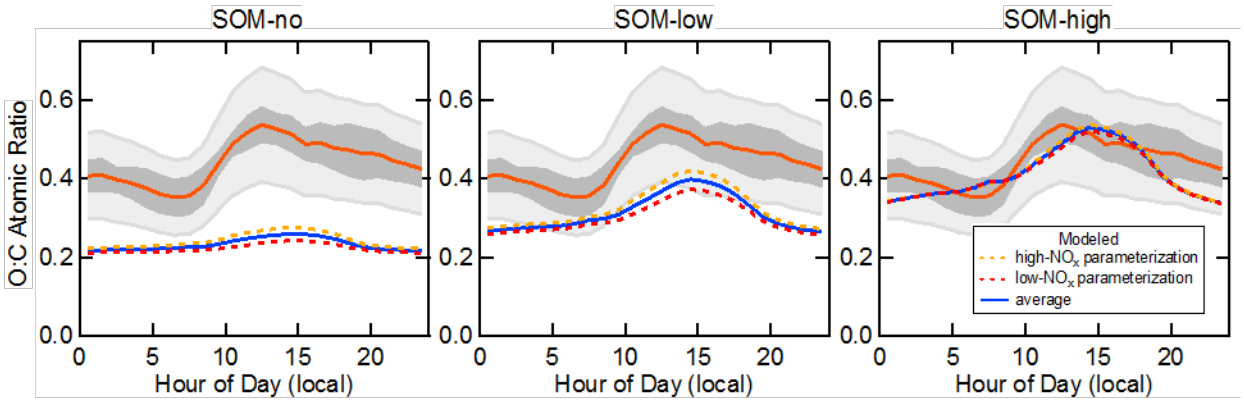
> There are some changes in the anthropogenic fraction of SOA when vapor wall losses are accounted for. *The anthropogenic fraction of SOA is defined here as the sum of the SOA from long alkanes and aromatics, which are emitted from combustion of fossil fuels, divided by the sum of the SOA from isoprene, monoterpenes and sesquiterpenes, which are emitted by trees, plants and other natural sources. The ^{14}C isotopic signature of fossil-derived VOCs is different from that of biogenically derived*

VOCs, and thus their respective contributions to SOA can be partially constrained via experimental analysis of the ^{14}C content of OA (Zotter et al., 2014). We assume the anthropogenic fraction is equivalent to the fossil fraction of SOA (termed $F_{SOA,fossil}$).

8. Page 30105: Since the diurnal variation of NOx concentrations haven't been taken into account in these simulations, would you expect the model to reproduce diurnal profiles well?

This question is in reference to the diurnal behavior of the O:C values that are discussed on Page 30105. The O:C of SOA shows only a minor dependence on the NO_x condition (Chhabra et al., 2010; Chhabra et al., 2011; Cappa et al., 2013), and thus we expect that the simulations are able to capture the general diurnal variations, which are driven mostly by diurnal changes in the particular SOA source and the POA contribution, even though the NO_x variability is not explicitly accounted for. However, since the absolute amount of SOA varies between the NO_x parameterizations, the SOA/OA ratio will differ between the low-NO_x and high-NO_x parameterizations for a given vapor wall loss condition (SOM-no, SOM-low or SOM-high). This could in turn influence the simulated O:C. Therefore, we have extracted the O:C ratios for the different NO_x parameterizations for each vapor wall loss condition and compared them to the average values, which are shown in the manuscript. The figure below shows the results of this exercise. It is apparent that the O:C differs only be a minor amount between the different NO_x parameterizations for a given vapor wall loss condition. The much larger influence comes from the different vapor wall loss parameterizations. Therefore, we can conclude that to a large extent not having accounted explicitly for diurnal variations in NO_x have minimal influence on the conclusions regarding the atomic ratios. We will add the following sentence to the manuscript:

The simulated diurnal profiles for a given vapor wall loss condition exhibit only very minor differences between the different NO_x parameterizations, with the high-NO_x parameterization giving a slightly higher O:C than the low-NO_x parameterization.



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