

Response to Interactive comment by Reviewer #1 on “Simulating Secondary Organic Aerosol in a Regional Air Quality Model Using the Statistical Oxidation Model: 2. Assessing the Influence of Vapor Wall Losses” by C. D. Cappa et al.

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

The authors have evaluated the performance of the Statistical Oxidation Model (SOM) within the UCD/CIT regional model for the South Coast Air Basin and Eastern US. The version of SOM used here was fit to laboratory chamber data after accounting for vapor wall losses. Low and high values of wall loss rates were considered to approximately account for the uncertainty in this process. Predicted SOA mass concentrations using the “high wall loss” fits are found to be in much better agreement with observations compared to “no wall loss” and “low wall loss” fits. The results are very interesting, the paper is well written and is recommended for publication in ACP after addressing the following comments.

We thank the reviewer for the comments, and address them each in turn below.

(1) The dependence of SOA yields on pre-existing aerosol surface area in the chamber makes it clear that vapor wall loss must be accounted in the interpretation of laboratory chamber data. However, it is not clear how the exercise of accounting for vapor wall loss yields a unique set of fitted values for k_{wall} , gas-phase yields of species with different volatilities (C^*), etc.

It is important to clarify here that the k_{wall} values used in this work were not derived as part of this work: the determination of k_{wall} values that are reasonable and appropriate for the Caltech chamber is discussed extensively in [Zhang et al., 2014]. In Zhang et al. [2014], an optimal value of k_{wall} was determined via simultaneous fitting of SOA formation experiments performed using different seed aerosol concentrations but with all other experimental conditions held constant. This exercise resulted in an optimal value of $2.5 \times 10^{-4} \text{ s}^{-1}$. This is equal to the “high” wall loss case used here. The “low” case was selected as a reasonable lower value to examine the sensitivity of the results to use of a lower k_{wall} value, i.e. to slower vapor wall loss. In Zhang et al. [2014], unique sets of SOM parameters (the fragmentation and functionalization parameters and the volatility decrease per oxygen atom added) were obtained for each k_{wall} assumed. In other words, the specific model parameters (“fitted values”) are specific to an assumed k_{wall} value. We have aimed to clarify this in the revised manuscript. Specifically, we have added the following in Section 3.3.1 (new text in italics):

“A base case set of parameters with no vapor wall losses assumed during fitting (termed SOM-no) was determined using $k_{\text{wall}} = 0$. In Zhang et al. [2014], an optimal value of $k_{\text{wall}} = 2 \times 10^{-4} \text{ s}^{-1}$ was determined for the California Institute of Technology chamber based on simultaneous fitting of the SOM to a set of toluene photooxidation experiments conducted at different seed particle concentrations. Unlike in Zhang et al. (2014), the values of k_{wall} used here were not determined during model fitting. This is because the absolute value of k_{wall} is not well constrained by a single experiment, and the simulations require vapor wall loss corrected parameters for VOCs besides toluene. Therefore, two specific bounding cases that account for vapor wall loss are instead considered based on the results from Zhang et al. (2014). Specifically, values of $k_{\text{wall}} = 1 \times 10^{-4} \text{ s}^{-1}$ and $2.5 \times 10^{-4} \text{ s}^{-1}$ are considered, corresponding to a low vapor wall loss case (SOM-low) and high vapor wall loss case (SOM-high), respectively.”

(2) Were the model fittings done at the end of each chamber experiment or as a function of time in a given experiment?

The model was fit as a function of time, as described in Zhang et al. [2014] and Cappa et al. [2013]. The SOM fits are not simply end of experiment fits. We have clarified this point in the current manuscript as follows (new text in italics):

In Section 3.2: “The parameters used in the current work have been determined by fitting to *time-dependent* data from SOA formation experiments conducted in the Caltech chamber both with and without accounting for vapor wall losses during the fitting process (discussed further below).”

and

In Section 3.3.1: “SOM was fit to *time-dependent* SOA formation experiments conducted in the California Institute of Technology chamber, following the methodologies described in Cappa et al. [2013] and Zhang et al. [2014].”

(3) It is stated that mass accommodation coefficient (α_{particle}) was assumed equal to 1. This indeed seems too conservative, especially if the SOA particles are semisolid and the gas-particle partitioning timescale is longer than currently assumed. If the chamber experiments were conducted under low RH then it is likely that the SOA particles were viscous semisolids. Please state the RH at which the fits were done and comment on how might the results change if $\alpha_{\text{particles}} < 0.1$.

The reviewer raises a good point. All experiments were done at low (<10%) relative humidity. And, in fact, in Zhang et al. [2014] we found that the effective mass accommodation coefficient was < 0.1 (specifically, $\sim 1-2 \times 10^{-3}$). As was shown in that work, if the same k_{wall} is used (i.e. $k_{\text{wall}} = 2.5 \times 10^{-4} \text{ s}^{-1}$) but α_{particle} was instead assumed = 1 (i.e. instantaneous partitioning), the magnitude of influence of vapor wall losses on the SOA may be less than or similar to when a smaller α_{particle} is assumed. In other words, it is not straightforward to quantitatively understand the impact of the $\alpha_{\text{particle}} = 1$ assumption used here compared to if some smaller α_{particle} had been used. Nonetheless, in all likelihood had a smaller α_{particle} been assumed then the impact of accounting for vapor wall losses on the simulated SOA concentrations would likely have been as large or larger. We now address this point in the conclusions through the addition of the following (new text in italics):

“Overall, the generally improved model performance when vapor wall losses are accounted for—in terms of both absolute and relative concentrations and in terms of SOA properties—suggests that accounting for this chamber effect in atmospheric simulations of SOA is important, although certainly requiring further examination. *For example, it was assumed here that the gas-particle mass accommodation coefficient was unity; had smaller values of α_{particle} been assumed during development of the SOM parameterizations used here it is likely that the increase in the simulated SOA concentration when vapor wall losses were accounted for would have increased for a given k_{wall} [Zhang et al., 2014].* Our results qualitatively agree with other recent efforts to assess the influence of vapor wall losses on ambient SOA concentrations [Baker et al., 2015; Hayes et al., 2015], but as our accounting for vapor wall loss is inherent in the SOA parameterization the simulations here serve to provide a more robust assessment.”

A similar question was raised by Reviewer #2, and we refer the reader to our Response to Point 2 to Reviewer #2 for additional discussion.

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