

Reviewer3

Interactive comment on “Modeling of Gas-Wall Partitioning of Organic Compounds Using a Quantitative Structure-Activity Relationship” by Sanghee Han, Myoseon Jang*, and Huanhuan Jiang

Department of Environmental Engineering Science, University of Florida, Gainesville, Florida, USA

mjang@ufl.edu

We thank reviewer 3 for the valuable comments on the manuscript.

Overall comment:

The authors report on a study that aims to allow for prediction of the loss of organic vapors to the walls of Teflon chambers based on the compound identity. This is an interesting framing of a complex problem. However, I unfortunately find that there are fundamental flaws with this study that I do not think can be addressed through revision. The two of these that I think are most important are: (i) the fundamental assumption that vapors partition only to organic matter that is bound to the walls of the Teflon chamber, rather than to the chamber itself, and (ii) the quality of the data shown does not appear sufficiently high to allow for robust determination of the desired properties, and there are issues with the overall experimental design. A third issue, although one that could be addressed through revision, is that I too often found the study details and the description of the procedures to be lacking in terms of clarity. After writing my review, I read that of Reviewer #1 and we seem to come to similar conclusions. I unfortunately do not see a path forward for this manuscript.

I find that the details provided regarding determination of the model parameters are insufficient to truly allow for this study to be reproduced. I think the authors need to do a more thorough job explaining how the calculations work and what assumptions go into them. I do not find it clear how the authors determined the $K_{w,i}$ values. They give a bunch of equations, but how this is determined experimentally is not clear to me. The “polynomial equation” (L179) that is used is not clear, and it is also not clear that this is really an observed value, versus a calculated value, given that the determination relies on calculated values of a variety of parameters. This, to me, makes it circular to compare the $K_{w,i}$ to the $1/C_i^*$ values (L249). Perhaps I am simply missing the distinction, but I do not find that the authors have presented their analysis in a sufficiently clear way to understand the details.

Summary of Response to Reviewer 3:

Based on the valuable comments from the reviewer, this manuscript was significantly improved. In summary

- 1) The gas-wall partitioning (GWP) of SVOCs is assumed as absorption-desorption kinetics to organic layer on the Teflon surface (OM_{wall}). This assumption set based on our

measurement of extracted OM_{wall} . The estimated composition and water contents of OM_{wall} using FTIR spectrum supports that the OM_{wall} is wax-like hydrophobic compounds (response to comment 1) which can dominantly impact on the GWP.

2) Model uncertainty

The uncertainties of predicted $K_{w,i}$ and $\alpha_{w,i}$ for each species were calculated based on the 95% confidence level boundary of each coefficient and the uncertainties of each physicochemical parameter in the polynomial equation to predict $K_{w,i}$ and $\alpha_{w,i}$. The uncertainties were presented with the predicted value on Table R2. Table 2 is updated with uncertainties in the revised manuscript.

3) Data uncertainty

The uncertainties of experimental data points in Fig. 3 of the revised manuscript were determined with a propagation error based on the quantitative procedure. The error associated with $C_{g,i}$ ranges 10-30% .

4) Procedure to estimate OM_{wall} composition

To response to the comment from the reviewer, the additional characterization of the organic layer (OM_{wall}) on the Teflon film surface of the chamber wall and the unused Teflon film was performed by using FTIR spectra.

The detail responses to the comments from Reviewer 3 are following:

Specific comments:

Comment 1) L47: I am concerned that this statement, which discusses partitioning into “absorbing organic matter on the Teflon film,” demonstrates a misunderstanding of how others have been thinking about the partitioning process. There is clear evidence that the partitioning occurs into the Teflon material directly, without need for any “organic matter.” Unless, of course, the authors are referring to the Teflon as “absorbing organic matter.” However, by stating that the issue is partitioning into OM that is “on” the Teflon film, it makes it seem as if the authors believe the key issue to be bound OM and not the Teflon material. Adsorbed OM can contribute to the partitioning, but is not necessary. Given the discussion in Section 2.3, where the authors sample OM from the Teflon walls, it seems clear that they are considering only partitioning into wall-bound OM. But this is a small fraction of the apparent wall mass, as used by Matsunaga and Ziemann (2010), that is important to consider. This issue is also raised by Reviewer #1 and is a core limitation of the current study.

Response:

The quantity and chemical properties of OM_{wall} varied between laboratories as summarized in Table R1 below:

Table R1. Summarized the assumption of the GWP on several previous studies.

Publication	Assumption	Chemical property of the surface layer	MW of wall material (g/mol)	chamber style (indoor/outdoor)
(Matsunaga and Ziemann, 2010;Krechmer et al., 2016)	Sorption mechanism (Eyring Hole theory) of SVOCs to the Teflon surface layer with effective mass concentration of organic aerosol particles (C_w) ^{a)} .	Teflon surface with C_w (absorbed SOA products)	200/250	Indoor
(Huang et al., 2018)	Two layers consisting of the surface Teflon layer and inner Teflon layer.	Surface Teflon layer with C_w (absorbed SOA products)	200	Indoor
This study	Absorption of SVOCs to the viscous organic layer on the Teflon film wall.	Wax-like organic matter	273	Outdoor

^{a)} The equivalent mass concentration (C_w) reported by Krechmer et al. (2016) is described as the absorbed SOA products on the Teflon surface by usage of the chamber. It is theoretically calculated based on the chamber history.

In this study, we assumed that organic vapor partition on the viscous organic layer on the Teflon film. This assumption was established based on our measurement of the chemical property of the solvent-extractable organic matter on the Teflon film wall. As seen the response to comment 4 from reviewer 1, the hydrophobic Teflon film favors the adsorption of wax-like organics in ambient air. Wax-like carbons were observed even in unused Teflon film by using FTIR. With the assumption of homogeneous coating of the Teflon film with OM_{wall} , the estimated thickness of the wax-like materials ranges 7.5-10.0 nm (calculated with the extracted OM mass (~800 μ g) from the film surface area (~1200 cm^2)). This thickness suggests multi-layer coating. The organic layer suggested by Krechmer et al. (2016) was a monolayer (1.5 nm or smaller) based on their theoretical assumption.

Based on our FTIR study (Fig. R3 in the response to comment 4 from reviewer 1), the estimated molecular weight of OM_{wall} is about 273 g/mol. The hygroscopicity of the Teflon film (Fig. S3), which is measured using FTIR data, suggests that the film surface material is hydrophobic. FTIR data shows that the water content associated with the film dramatically changes at the RH higher than 0.7. This viscous and hydrophobic OM_{wall} layer can impact on GWP. Shiraiwa et al. (2011) reported that the diffusivity of atmospheric chemical species in semisolid aerosol materials is very slow with a time scale ranging from several hours to days. The wax-like material determined by our study may also penetrate the certain layers of the Teflon film near the surface and influence the property of the Teflon film surface.

In the current knowledge, the understanding of the characteristics of OM_{wall} is uncertain. The molecular weight of OM_{wall} in the previous studies was determined by using the average molecular weight of SOA based on the value in the literature. For example, Matsunaga and Ziemann (2010) and Krechmer et al. (2016) determined the OM_{wall} 's molecular weight based on SOA studies reported by Seinfeld et al. (2001). In their studies, the OM_{wall} molecular weight was not determined theoretically or by measuring of the chemical composition of Teflon surface matter. In the study by Krechmer et al. (2016), the OM_{wall} molecular weight was 250 g/mol, which is the same molecular weight with a FEP monomer. However, the actual molecular weight of FEP ranges 76,000-603,000 (Wypych, 2016). Furthermore, the monomeric

FEP with 250 g/mol molecular weight is very volatile (order of 10^3 mmHg) because Teflon functionality significantly reduces volatility. Thus, the monomeric FEP are gaseous and it cannot be regarded as the molecular weight of a surface material. Thus, the proposed molecular weight (200-250 g/mol) in the previous studies are uncertain. Importantly, either wax-like matter of this study or the short chain-length Teflon layer used in other studies are viscous and they will slow down the diffusion of organic molecules. Our QSAR approach to produce predictive polynomial equations for GWP is developed based on the physicochemical properties of organic compounds and will not be influenced by any assumption for OM_{wall} at given A/V and MW_{OM} .

Comment 2) L58: it is unclear why this should be the mean molecular weight of the organic material on the Teflon. The MW term is needed to convert from vapor pressure to saturation concentration, and thus should be the MW of the SVOC under consideration.

Response:

Please find the response to comment 1.

Based on our determined composition, the vapor pressure of an organic compound is in the order of 10^{-9} mmHg, suggesting that this compound is nearly nonvolatile.

Comment 3) L62: Differences in O:C ratios does not provide support for there being “diverse functionalities.” If the focus is on functionalities, then I suggest the authors focus on functionalities and not generic properties such as O:C.

Response:

The sentence pointed by the reviewer has been revised and reads now,

“SOA products originating from the oxidation of hydrocarbons are diverse in functionalities, such as alkanes, aldehyde, carboxylic acids, ketones, and alcohols. The polarity of SOA is closely related to the oxygen to carbon (O:C) ratio of SOA. For example, the O:C ratio of α -pinene SOA is 0.43 on average (Zhang et al., 2015; Chen et al., 2011) but that of isoprene products is about 0.8 (Bertram et al., 2011; Chen et al., 2011; Kuwata et al., 2013).”

Comment 4) L68: The definition of $\alpha_{w,i}$ does not seem correct to me. It is the mass accommodation coefficient to the wall, not “the fraction that reversible uptake of a gas-phase species will occur upon collision with the chamber wall.”

Response:

This sentence has been removed

Comment 5) L96: It is not clear how the procedure the authors use “reduce[s] the delay attributable to inject chemicals.” Reduces how and from what? The mixing time reported is still 10 minutes, which Krechmer et al. (2016), among others, has shown to be a very long time compared to the time to partition to walls, especially in a highly turbulent chamber as would be the case here based on the procedure described.

Response:

The summary table for the injection of organic vapor into the chamber is shown in the response to comment 1 from reviewer 1 (Please find the Table R1 in the response to comment 1 from reviewer 1). In order to avoid the potential condensation reactions between different chemical functionalities, each class of organic compounds was separately vaporized into the chamber. With the three groups, the injection of 21 compounds into the East chamber takes at least 30 minutes. UF-APHOR is a dual chamber system with the air tighten door in the middle. The organic vapor introduced into the East chamber was transferred to the West for 10 minutes by opening the middle door.

Comment 6) L104: The authors state “to ensure particle formation. . .”. Was there particle formation and was this desired? This does not seem correct.

Response:

Thank you for pointing this sentence. This has been corrected to provide our original intention to readers and reads now,

“No particle appeared after vaporizing organic chemicals into the chamber air based on particle data, which was monitored using a scanning mobility particle sizer (SMPS, TSI 3080, Shoreview, MN, USA) and a condensation particle counter (CPC, TSI 3022A, Shoreview, MN, USA).”

Comment 7) L102&108: How long were samples collected on the denuders?

Response:

It is mentioned in the SI section 1. The sampling duration for the first sample was 15 minutes and was gradually increased up to 35-40 minutes.

Comment 8) L109: Based on the data presented, the measurement uncertainties are undoubtedly underestimated here. It would be useful to have a more accurate representation of the true uncertainty.

Response:

The information of the error associated with the organic vapor concentration shown in Fig. 3 was updated and included in the figure caption. The concentration of gas phase SVOCs are calculated by the equation below:

$$C_{g,i} = \frac{f\left(\frac{\text{peak area of target species}}{\text{peak area of internal standard}}\right) \times \text{dilution factor}}{\text{Flow rate} \times \text{sampling time}}$$

$f(x)$ is the calibration curve for each compound obtained from external standard. The error was estimated with a propagation error calculation method. Thus, the calculated uncertainty of data points includes flow rate, sampling time, GC measurement, and the uncertainty of the GC calibration curve. The calculated uncertainty ranges 10-30% of the concentration and it is

updated on the Fig. 3 and the caption of Fig. 3 as follows:

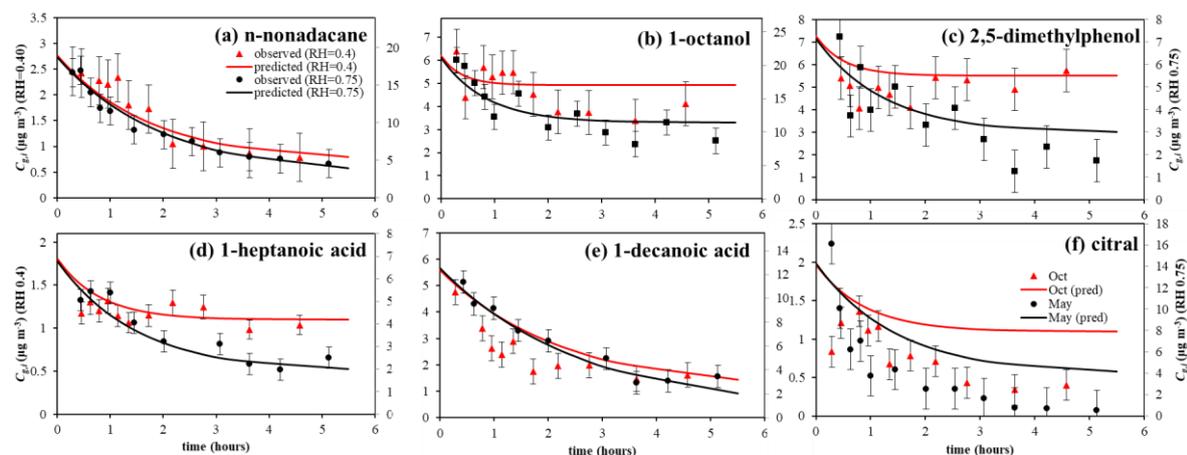


Figure 3. The observed concentrations of the gaseous SVOCs ($C_{g,i}$, plot) in the chamber air for the two experiments at two different humidity (October 8, 2018 at RH=0.40 and May 15, 2018 at RH= 0.75) and predicted $C_{g,i}$ (line) as a time series. $C_{g,i}$ was calculated by the equation ($C_{g,i} = \frac{f(\frac{\text{peak area of target species}}{\text{peak area of internal standard}}) \times \text{dilution factor}}{\text{Flow rate} \times \text{sampling time}}$). The error bar associate with $C_{g,i}$ estimated using propagation error based on the quantitating procedure of GC/MS data ranges 10–30% of the estimated concentrations.

Comment 9) Section 2.1: The authors never directly state what chemicals they actually use. This would seem the place to give this detail.

Response:

Table 1 in the main manuscript and the section S1 in supporting materials provide the chemical information used in this study.

Comment 10) Section 2.3: there is no mention of the RH at which the measurements are made. Are they really made at 10% and 80% RH? How was the RH controlled, if this is the case? How long were samples allowed to equilibrate? This is critical information if the statements regarding the wall-bound OM hygroscopicity are to be believed on L140.

Response:

The detail information of the experimental procedure is described in the Hygroscopicity of OM_{wall} in Section S2. In order to respond to the reviewer, we added the information about humidity change rate into Section S2 in the revised SI.

“Relative humidity increased to 0.8, stands for 15 minutes, and decreased at 0.01/minutes. Hygroscopicity experiments using FTIR were repeated three times.”

Comment 11) L166: it is unclear to me how this statement is true. If the $k_{on,i}$ increases the $k_{off,i}$ will increase the balance and give the same $K_{w,i}$, which is an equilibrium property.

Response:

This has been corrected.

“The larger A/V ratio can yield the larger $k_{on,i}$, and thus $k_{off,i}$, at a given $K_{w,i}$ (Eq. 7), ultimately leading the shorter GWP equilibrium time (see Sect. 3.2).”

Comment 12) L176: is unclear why the $\alpha_{w,i}$ would depend on the chamber area to volume ratio. Alpha is simply a property of the species condensing and material onto which it is condensing.

Response:

This has been corrected and reads now,

“The prediction of $k_{on,i}$, which is dependent on $\alpha_{w,i}$ and the chamber’s characteristics (A/V). The $\alpha_{w,i}$, diverged with the physicochemical properties of SVOC can also be predicted by using QSAR approach as follows:”

Comment 13) Eq. 13: It is unclear how Eq. 13 derives from Eq. 5. This needs to be shown more explicitly.

Response:

The coefficient in the exponential term in Eq. 5 is a rate constant consisting of $k_{on,i}$ and $k_{off,i}$. The characteristic time is inversely related to the rate constant.

Comment 14) L187: I disagree with the authors contend that the change in the functional group distribution is small year around. They show a total of 2 spectra. The spectra actually show substantial differences. Further, the authors have not quantified any of the differences, taking only a very qualitative approach period if they wish to make this type of statement they need to support their conclusions more with their measurements.

Response:

- a. Difference between two FTIR spectra: Based on the chamber log, high concentration of inorganic seed was injected before the OM_{wall} extraction on 03/14/2019. The FTIR spectrum measured on 03/14/2019 might affected by inorganic seed. Thus, additional extracted OM_{wall} from chamber have been analyzed and shown in Fig. R3 for the response to comment 4 from Reviewer 1. Fig. S4 is replaced with Fig. R3 for the response to comment 4 from Reviewer 1.
- b. Quantification of the FTIR spectrum: In order to response to the reviewer, Fig. 1 and Fig. S4 are updated the detail information was added below Table S1.

“The procedure of least-squares curve fitting was implemented for the multi-component analysis. The fitting parameters include the center frequency, the peak absorbance, and the half width at half-height. The band shapes in the FTIR absorbance spectrum were approximated by a Gaussian function (Li et al., 2016;Jang and Kamens, 2001;Jang et al., 2008). The decoupled the FTIR bend for each functional group was applied to estimate the functionality composition of OM_{wall} using the relative intensity of the functional group determined from various reference compounds.”

Comment 15) Fig. 3: The data, to me, do not appear of sufficiently high quality to allow for

robust determination of the model parameters derived by the authors. In many, or even most, cases the fits to the observations appear poor. I strongly suggest that a rigorous discussion of the measurement uncertainties and data quality, along with the fit quality, is required for this work to be publishable.

Response:

Please find the response to comment 8.

The uncertainty of experimentally measured $C_{g,i}$ was updated on Fig. 3 and the information of the uncertainty of measurement data was updated on the figure caption.

Comment 16) Section 4.3: A discussion of uncertainties is lacking entirely. The authors give no sense of whether the (for example) difference of a factor of 7 in $K_{w,i}$ for 1-decanoic acid and *n*-nonadecane is statistically justified.

Response:

In order to response to the reviewer, the model uncertainty analysis was performed for $K_{w,i}$ and $\alpha_{w,i}$. The uncertainties of predicted $K_{w,i}$ and $\alpha_{w,i}$ were determined from the standard error associating with the final polynomial equations (Eq. 14 and Eq. 15, respectively) and uncertainty of each physicochemical descriptor.

The discussion in the Section 4.3 has been revised based on the updated polynomial equation and uncertainty analysis and reads now,

“For example, 1-decanoic acid’s $K_{w,i}$ is nearly 4 times higher (18.72 ± 6.6) than *n*-nonadecane’s $K_{w,i}$ (5.34 ± 2.5), although 1-decanoic acid’s estimated $p_{L,i}^{\circ}$ (1.18×10^{-3} mmHg) is close to *n*-nonadecane’s $p_{L,i}^{\circ}$ (1.09×10^{-3} mmHg). In general, a longer τ_{GWP} was found for SVOCs with high $K_{w,i}$ and low $\alpha_{w,i}$. For example, *n*-eicosane’s τ_{GWP} (127 min) was significantly higher than that of 2-heptanol (30 min), while *n*-eicosane and 2-heptanol had a similar $\alpha_{w,i}$ ($2.9 (\pm 0.2) \times 10^{-6}$ and $3.3 (\pm 0.1) \times 10^{-6}$, respectively). The dissimilar $K_{w,i}$ values of *n*-eicosane and 2-heptanol (7.49 ± 3.5 and 0.44 ± 0.2 , respectively) occurred with large differences in τ_{GWP} .”

In addition, Table 2 in the manuscript is updated with uncertainties.

Table R2. Chemicals injected into the chamber and their physicochemical parameters (at 298 K) predicted using PaDel-Descriptor and estimated $K_{w,i}$, $\alpha_{w,i}$, and τ_{GWP} under a given condition (298K, RH=0.75).

No.	Chemical	MW	Molecular descriptors ^{a)}					p_L° ^{b)}	$K_{w,i} \pm unc^c)$	$\alpha_{w,i} \pm unc^c)$ ($\times 10^{-6}$)	τ_{GWP} (s)
			$H_{d,i}$	$H_{a,i}$	E_i	S_i	α_i				
1	1-hexanoic acid	116	0.59	0.41	0.15	0.51	14.08	0.078	1.27±0.5	2.7±0.2	3671
2	1-heptanoic acid	130	0.59	0.41	0.15	0.51	16.26	0.026	2.64±1.0	2.5±0.1	5185
3	1-octanoic acid	144	0.59	0.41	0.15	0.51	18.45	0.009	5.33±2.0	2.3±0.1	6563
4	1-decanoic acid	172	0.59	0.41	0.15	0.51	22.82	0.001	18.72±6.6	1.9±0.1	8832
5	Benzoic acid	122	0.59	0.46	0.78	0.93	7.52	0.020	13.39±4.9	2.2±0.1	7097
6	Pyruvic acid	88	0.43	0.68	0.29	0.88	8.48	0.256	1.69±0.6	1.9±0.1	4661
7	2-heptanol	116	0.35	0.40	0.19	0.41	17.49	0.317	0.44±0.2	3.3±0.1	1799
8	1-octanol	130	0.35	0.39	0.21	0.45	19.68	0.109	0.86±0.4	3.0±0.2	2975
9	1-nonanol	144	0.35	0.39	0.21	0.45	21.86	0.036	1.82±0.8	2.7±0.2	4499
10	Benzyl alcohol	108	0.35	0.56	0.83	0.88	8.75	0.093	5.85±2.4	2.4±0.1	5904
11	Phenol	94	0.55	0.43	0.83	0.88	6.56	0.988	0.38±0.2	2.6±0.2	1735
12	2,5-dimethylphenol	122	0.55	0.43	0.85	0.83	10.93	0.123	1.44±0.6	2.3±0.1	4352
13	2,6-dimethoxyphenol	154	0.04	0.284	0.74	1.36	14.76	0.041	11.45±6.0	2.2±0.1	7458
14	n-heptadecane	240	0.00	0.08	0.04	0.13	39.36	0.005	2.43±1.1	3.8±0.3	4726
15	n-nonadecane	308	0.00	0.08	0.04	0.13	43.73	0.001	5.34±2.5	3.2±0.2	6758
16	n-eicosane	324	0.00	0.08	0.04	0.13	45.91	0.001	7.49±3.5	2.9±0.2	7629
17	2-Dodecanone	184	0.00	0.42	0.18	0.60	27.19	0.053	1.69±0.8	2.5±0.1	4982
18	2-Tridecanone	198	0.00	0.42	0.18	0.60	29.38	0.022	2.74±1.3	2.3±0.1	6314
19	Decanal	156	0.00	0.42	0.15	0.51	22.82	0.193	0.97±0.5	2.9±0.2	3390
20	Citral	152	0.00	0.46	0.15	0.51	18.45	0.142	2.66±1.3	2.8±0.2	5032
21	Benzaldehyde	106	0.00	0.47	0.15	0.51	7.52	0.982	2.26±1.1	3.3±0.2	3995

^{a)} Based on the QSAR approach with PaDEL-Descriptor (Yap, 2011).

^{b)} Calculated through group contribution (Zhao et al., 1999; Stein and Brown, 1994).

^{c)} The uncertainty propagation error calculated based on the 95% confidence level boundary of predictive polynomial equations for $K_{w,i}$ and $\alpha_{w,i}$.

Comment 17) L20: I suggest that the first sentence be rewritten. As currently written, it is not a fully formed sentence. What does “atmospheric process of reactive hydrocarbons” mean? I find this ambiguous and difficult to parse.

Response:

The sentence pointed by the reviewer has been revised and reads now,

“Organic Aerosol (OA) consists of primary sources originating from fuel combustion, industries, and vehicles and Secondary Organic Aerosol (SOA) which forms via the atmospheric reactions of reactive hydrocarbons with atmospheric oxidants (i.e., an OH radical, ozone, and a nitrate radical).”

Comment 18) L23: SOA can constitute much more than 40% of the OA budget in a region. “Up to 40%” is not correct.

Response:

The sentence pointed by the reviewer has been revised and reads now,

“SOA constitutes a large proportion (ranging from 20% to 90%) of OA in the ambient air (Hallquist et al., 2009) and it can significantly impact on climate (Seinfeld and Pandis, 2016), visibility (Park et al., 2003), and human health (Cohen et al., 2017).”

Comment 19) L26: technically, many SOA models are not mass-conserving, and thus do not take a “mass balance” approach.

Response:

This sentence has been removed.

Comment 20) L38: To what, more specifically, does “the gas-wall process” refer? Conventionally, people have used GWP (in this context) to mean gas-wall partitioning, not process.

Response:

The definition of GWP has been changed from the gas-wall process to gas-wall partitioning.

Comment 21) L42: “underrated” should be “underestimated” or “under-predicted”.

Response:

This has been corrected and reads now,

“La et al. (2016) indicated that the SOA yield inferred from a chamber study can be under-predicted by more than 50% for alkane and alkene series.”

Comment 22) L44: I suggest these two sentences need be rewritten. They are very difficult to understand, yet I am also not certain that they are correct. What “database?” “Burdensome” how?

Response:

The sentence pointed by the reviewer has been revised and reads now,

“To date, the predictive model to account for the impact of GWP on SOA yields remains controversial due to the limitations of the experimental approach to measure the vapor concentration of organic species. For example, the actual vapor concentration of the organic compound in the initial point is burdensome because the experimental procedure required for the vaporization of organic species into a chamber delays the measurement of the initial concentration of organic vapor without GWP.”

Comment 23) L52: The citation of Im et al. (2014) here seems unnecessary and arbitrary.

Response:

This has been removed.

References:

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