

We appreciate the reviewer for providing us useful comments. In the following, original reviewer comments, author's responses, and corresponding updates on the main text are shown as purple, black, and *italic*. Line numbers in the responses correspond to those in last submitted version.

(1.1) Overall the authors have responded to the referee comments very well. I have just one serious concern, which is that the discussion, data and analysis around uncertainties in using 3 hour data for $C_{\text{surface}}/C_{\text{gas}}$ is confusing and lacking (detailed below). I recommend publication subject to this minor revision.

We thank the reviewer for the positive comment on our last reply. Response to the specific comment is given below.

(1.2) Specifically, Text S1 indicates that uncertainties are potentially major. This needs stating and discussing (including implications for results) explicitly in the results section, rather than referring readers to the supplementary section without further discussion. In addition, Text S1 makes very little sense to me: Figure S4b does not exist. I don't know what is overestimated by 7 and 55 %, and what the implications are for the main paper results. And the last two sentences of Text S1 makes no sense to me: what is the method that is doing the overestimating, what does employing 10 times mean, and what are the implications for main text results?

We thank the reviewer for this comment. The original content of Text S1 described the discrepancy between the K_{eq} and our approximation of K_{eq} as $1/[C_{\text{gas}}/C_0]_{\text{at 3 hours}} - 1$. We acknowledge the reviewer for pointing out that the figure number in the previous manuscript was inaccurate. The figure number was updated in the revised manuscript.

In the revised manuscript, we fully updated the corresponding descriptions, considering the reviewers' comment. Namely, we provided a detailed comparison of K_{eq} and $1/[C_{\text{gas}}/C_0]_{\text{at 3 hours}} - 1$ for the room temperature experiments in Table S3. The employment of the room temperature data provides a quantitative comparison, as any temperature controlling processes were not needed. The experiments at room temperature were conducted for three times, allowing to estimate experimental uncertainties as standard deviations among the replicated runs. The result is summarized in Table S3 of the revised supplement file, demonstrating that the K_{eq} and $1/[C_{\text{gas}}/C_0]_{\text{at 3 hours}} - 1$ agree well within experimental uncertainties. Overall, the approximation method, with an average overestimation by 22%, would not influence our main results. This result demonstrated the validity of employing $1/[C_{\text{gas}}/C_0]_{\text{at 3 hours}} - 1$. The corresponding descriptions were updated in the revised manuscript and supplement file, as detailed in the following.

In main text:

~~“Potential uncertainties associated with the employment of the data at 3 hours after injection as a proxy for gas-surface partitioning are summarized in Text S1.” (Lines 177-178)~~

“It is challenging to retrieve the value of $[C_{\text{surface}}/C_{\text{gas}}]_{eq}$ by fitting the data of the low-temperature experiments using the two-layer model, since the chamber was cooled after the injection of n-alkanes. Alternatively, the value of $[C_{\text{surface}}/C_{\text{gas}}]_{eq}$ was approximated using $1/[C_{\text{gas}}/C_0]_{\text{at 3 hours}} - 1$, assuming that diffusion of n-alkanes to the inner layer was still a minor loss process within 3 hours. Potential uncertainties associated with this approximation are summarized in Text S1. The uncertainties were estimated in two ways: (1) kinetic simulation based on fitting

parameters in Figure 3 (Figure S5) and (2) comparison of the retrieved values of $[C_{\text{surface}}/C_{\text{gas}}]_{\text{eq}}$ (i.e., K_{eq}) and $1/[C_{\text{gas}}/C_0]_{\text{at 3 hours}} - 1$ at room temperature (Table S3). The room-temperature experiments were conducted for three runs, allowing for the estimation of experimental uncertainties as standard deviation. Although the kinetic simulation implies overestimates of 7 - 55%, the measurement-based comparison demonstrates that $1/[C_{\text{gas}}/C_0]_{\text{at 3 hours}} - 1$ and $[C_{\text{surface}}/C_{\text{gas}}]_{\text{eq}}$ agreed within the experimental uncertainties, thereby supporting the validity of the approximation.” (Lines 199-201)

In supplement:

We revised the title of Figure S5 to clarify:

“(b) Time series of the ratio of mass in surface and gas phase ($C_{\text{surface}}/C_{\text{gas}}$) and the ratio of mass not in and in the gas phase ($1/[C_{\text{gas}}/C_0] - 1$) for C_{14} and C_{19} n-alkanes. Solid and dashed lines represent C_{14} and C_{19} n-alkanes respectively. Red and blue lines represent the values of $C_{\text{surface}}/C_{\text{gas}}$ and $1/[C_{\text{gas}}/C_0] - 1$ —~~theoretical and measured $C_{\text{surface}}/C_{\text{gas}}$ in the main text~~ respectively. The gray solid line indicates 3 hours as we choose the 3-hour measurements ($1/[C_{\text{gas}}/C_0]_{\text{at 3 hours}} - 1$) to ~~calculate $C_{\text{surface}}/C_{\text{gas}}$ approximate K_{eq} in the main text.~~”

Also, we added a comparison in Table S3:

“Table S3. Fitting and measurement results for room-temperature experiments.

Compound	Experiment shown in Figure 3 ^a			All three room-temperature experiments ^b		
	k_1 (s ⁻¹)	k_{-1} (s ⁻¹)	k_2 (s ⁻¹) ^c	K_{eq}	$1/[C_{\text{gas}}/C_0]_{\text{at 3 hours}} - 1$	relative difference
C_{14} n-alkane	2.76×10^{-4}	2.07×10^{-4}	9.39×10^{-6}	1.00 ± 0.27	1.14 ± 0.23	$16\% \pm 8\%$
C_{15} n-alkane	2.79×10^{-4}	2.07×10^{-4}	1.87×10^{-5}	1.08 ± 0.19	1.23 ± 0.21	$13\% \pm 6\%$
C_{16} n-alkane	3.41×10^{-4}	1.84×10^{-4}	2.67×10^{-5}	1.72 ± 0.17	1.98 ± 0.14	$16\% \pm 11\%$
C_{17} n-alkane	4.63×10^{-4}	1.82×10^{-4}	3.60×10^{-5}	2.73 ± 0.50	3.27 ± 0.15	$23\% \pm 16\%$
C_{18} n-alkane	6.35×10^{-4}	2.13×10^{-4}	4.35×10^{-5}	3.91 ± 1.27	4.85 ± 0.57	$31\% \pm 23\%$
C_{19} n-alkane	1.17×10^{-3}	2.25×10^{-4}	4.62×10^{-5}	8.69 ± 4.37	10.53 ± 2.80	$35\% \pm 27\%$

^a Optimized parameter sets of the two-layer model used in Figure 3. Best-fit parameters were obtained by the Newton method via Wolfram Mathematica 13.1.

^b (Mean value) \pm (standard deviation) are presented.

^c k_2 obtained here was not used in Section 3.3 (characterization of diffusion in the Teflon wall). These fittings overestimated the first-order loss rate constant for low volatile species, $C_{16} - C_{19}$ n-alkanes, as shown in Figure 3.”

And we fully revised Text S1:

“Text S1. Uncertainty in approximating K_{eq} as $1/[C_{\text{gas}}/C_0]_{\text{at 3 hours}} - 1$ — ~~$C_{\text{surface}}/C_{\text{gas}}$ by employment of 3-hour measurement data.~~”

“The uncertainties in approximating K_{eq} as $1/[C_{\text{gas}}/C_0]_{\text{at 3 hours}} - 1$ were estimated in two ways. First, the fitting parameters for room-temperature experiment in Figure 3 were used to simulate the kinetic process of wall loss (Figure S5a). Specifically, the values of $C_{\text{surface}}/C_{\text{gas}}$ and $1/[C_{\text{gas}}/C_0] - 1$ for C_{14} and C_{19} n-alkanes were retrieved, which were shown as red and blue lines in Figure S5b. These two n-alkanes were chosen, as they represent the highest and lowest volatile species in the room-temperature experiment. For both n-alkanes, $C_{\text{surface}}/C_{\text{gas}}$ stabilized by 3 hours, suggesting gas-surface partitioning reached equilibrium. In other words, K_{eq} equals to

$C_{\text{surface}}/C_{\text{gas}}$ at 3 hours. The discrepancy between the red lines and corresponding blue lines at 3 hours in Figure S5b was thus the bias caused by approximating K_{eq} as $1/[C_{\text{gas}}/C_0]_{\text{at 3 hours}} - 1$. For C_{14} and C_{19} n-alkanes, this approximation overestimated the values of K_{eq} by 7% and 55%, respectively.

The uncertainties were also estimated by the room-temperature experimental results. Since no temperature controlling process was needed for room temperature experiments, the experimental data can be fitted using the two-layer model. Values of k_1 and k_{-1} , and thus K_{eq} ($=k_1/k_{-1}$) can be obtained. Three sets of experiments were conducted at room temperature, which allows for estimating the experimental uncertainties as standard deviations among these replicated runs. As shown in Table S3, values of K_{eq} and $1/[C_{\text{gas}}/C_0]_{\text{at 3 hours}} - 1$ agree well within experimental uncertainties. On average, this approximation overestimates K_{eq} by 22%, which would not influence our main results.

In summary, although the model simulation implies potentially large uncertainties, the comparison based on experimental data demonstrates the validity of the approximation. ~~Uncertainties in $C_{\text{surface}}/C_{\text{gas}}$ for C_{14} and C_{19} n-alkanes at room temperature by employment of the 3-hour measurement data are shown in Figure S4b. The discrepancy between the red lines and corresponding blue lines at 3 hours is the bias caused by the employment of 3-hour measurements. For C_{14} and C_{19} n-alkanes, this method overestimates 7% and 55%. A more extreme case was used to simulate the low-temperature experiment by employing 10 times at k_1 of C_{19} n-alkane and the maximum k_2 in all the experiments. In this case, this method overestimates $C_{\text{surface}}/C_{\text{gas}}$ within 62% of the theoretical values.~~