Response to Anonymous Referee #2

We thank the reviewer for the constructive suggestions/comments. Below we provide a point-by-point response to individual comment (Reviewer comments and suggestions are in italics, responses and revisions are in plain font; revised parts in responses are marked with red color; page numbers refer to the modified AMTD version).

Comments and suggestions:

Overall Comments. In the work submitted, Li et al. presented the design, construction, and validation of a dynamic chamber system, which can be used to measure uptake coefficient on bulk solid-phase sample. Also, author have shown a relatively good agreement between flow tube, reference, and this study. The technique they developed is very important, and they also carried out tests for operational parameters and validation experiments very comprehensively. The paper is also well-written, and I recommend the manuscript to be published after corrections.

Responses and Revisions:

Thanks for the positive comments and feedback from the reviewer.

Comments and suggestions:

Specific Comments. Page 3 line 25-30: The detailed calculation for uptake coefficient has shown here. The wall-loss effect on the uptake coefficient determination is supposed to discussed in detail. Also, wall-lost correction is necessary to described in 2.1.1. section.

Responses and Revisions:

Thanks for the comments.

Following the reviewer's suggestion, we have added some detailed discussions in the revised manuscript (page 4, line 1-6), as shown below:

"…where C_{ref} (in molecules m^{-3}) is the gas reactant concentration measured at the outlet of a blank chamber prior to the uptake experiment. Since our chamber system had a dynamic flow-through feature, a constant rate of wall loss (once existed) was observed, i.e., the ratio of C_{ref} to C_{in} showed a fixed value during our uptake experiments (see Fig. 3). Thus, using C_{ref} (instead of C_{in}) for flux calculation already accounted for gas losses on chamber walls. Before each gas uptake experiment, C_{ref} was determined mimicking the chamber conditions (i.e., gas flow rate, gas mixing state,

Comments and suggestions:

Specific Comments. As author explained why outlet position C is used to represent the average concentration due to existence of vertical concentration gradient inside the chamber. However, the smaller horizontal concentration gradient is observed in Fig. 4 and Fig. S2 due to mixing fan. If another mixing fan is placed at both sides of chamber or buffer flask for completing mixing, the vertical concentration gradient might be minimized.

Responses and Revisions:

Thanks for the comments.

As the reviewer pointed out, the smaller horizontal concentration gradient observed in Fig. 4 and Fig. S2 should be due to the incomplete mixing. And vertical concentration gradient can also be minimized by adding a mixing fan at the other side of the chamber (close to the buffer flask on the chamber bottom). However, the related modification/improvement is changeling. The motor used to drive the fan needs to be fixed in a proper way on the bottom side of the chamber, which seems not easy for our current chamber setup. An alternative way may be adding another fan on the lid of the chamber or increasing the speed of the installed fan.

Comments and suggestions:

Specific Comments. Surface morphology of KI coating sample on Vt have shown no significant effects on the deposition velocity. If possible, different preparation on KI sample is better to use optical microscope or SEM/TEM to characterize their morphology.

Responses and Revisions:

Thanks for the suggestion.

As the reviewer suggested, we have added some photos showing the different surface morphologies of the KI coatings prepared via different procedures. Due to the complexity of the coating samples, it is difficult to characterize their morphology by optical microscope or SEM/TEM. The photos are therefore made by a mobile phone camera. The related modifications in the revised manuscript (page 14, line 17-19, and page 15 line 1-5) are shown below:

"… Figure 5 displays the four types of surface phase-state/morphology of the prepared KI coatings. Apparently, different preparation procedures generated distinct surface states/morphologies, especially among Type I, II/III and IV. Figure 6 shows the calculated V_t corresponding to these coating types. …
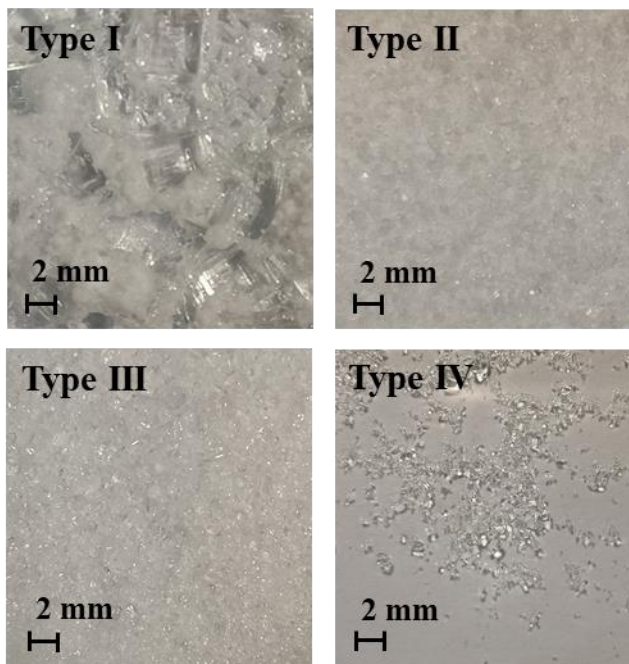

Figure 5. Characteristic morphologies of coated KI (top view) observed by a mobile phone camera. Type I: KI film; Type II: KI grain pre-humidified in a 50% RH N_2 environment for 30 min; Type III: KI grain pre-humidified in a 73% RH N_2 environment for 30 min; Type IV: saturated KI solution with deposited KI grains which cannot be further dissolved. Note that Type IV has a smooth liquid surface."

Comments and suggestions:

Specific Comments. Regarding to Fig. 7, there is a clear boundary for mass transport and surface reaction. Could author explain how to define "surface-reaction-limited", "transition regions", and "transport-limited region"

Responses and Revisions:

Thanks for the comments.

In our manuscript, Fig. 7 (i.e., Fig. 8 in the revised manuscript) is used to give a schematic overview on how an overall uptake is influenced by mass transport (R_t) and reactions on sample surfaces (R_s). For a multiphase reaction (i.e., uptake process), normally a gas reactant first needs

to be transported to the vicinity of a sample (solid or liquid phase), and then collides with the sample surface to trigger certain reactions. If a surface reaction has an extremely low rate (indicative of a very small γ, $R_t \ll R_s$), the overall uptake process is determined by the surface reaction (surface-reaction-limited region). On the other hand, if transport takes a very long time ($R_t \gg R_s$), the overall uptake will be limited by the transportation (transport-limited region). If these two processes have similar rates, both will play a critical role in determining the overall uptake (transition region). In Fig. 8, we artificially define the case when $R_t/R_s < 0.1$ (i.e., one order of magnitude lower) as the surface-reaction-limited region ($R_t \ll R_s$) and the case when $R_t/R_s > 10$ (i.e., one order of magnitude higher) as transport-limited region ($R_t \gg R_s$). And the case in between is defined as the transition region, where R_t is comparable to R_s.

We have added some explanations in the revised manuscript (page 17, Fig. 8 caption and page 18 line 11-17), as shown below:
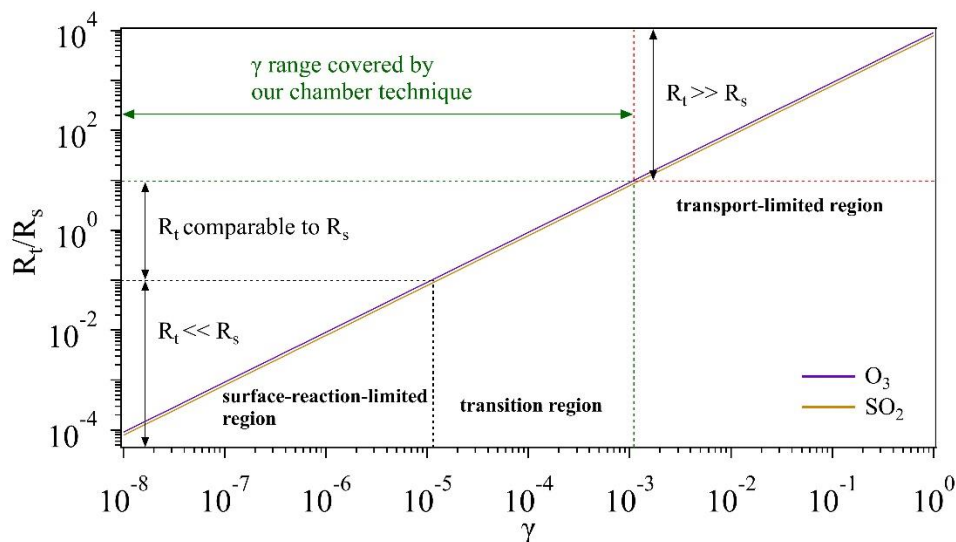
"

Figure 8. Schematic of different regions for O_3 and SO_2 uptake. Note that we artificially define the case when $R_t/R_s < 0.1$ (i.e., one order of magnitude lower) as the surface-reaction-limited region ($R_t \ll R_s$) and the case when $R_t/R_s > 10$ (i.e., one order of magnitude higher) as transport-limited region ($R_t \gg R_s$). And the case in between is defined as the transition region, where R_t is comparable to R_s. The transport resistance R_t is calculated based on the measured average V_t (Sect. 3.1.3). Calculations for the surface resistance R_s are referred to conditions of room temperature (296 K) and 1 atm.

4 Conclusions

… The chamber results agree well with those from the flow tube method and the literature data.

Figure 8 gives a schematic overview on how an overall uptake is influenced by mass transport (R_t) and reactions on sample surfaces (R_s). For a multiphase reaction (i.e., uptake process), normally a gas reactant first needs to be transported to the vicinity of a sample (solid or liquid phase), and then collides with the sample surface to trigger certain reactions. If a surface reaction has an extremely low rate (indicative of a very small γ, $R_t \ll R_s$), the overall uptake process is determined by the surface reaction (surface-reaction-limited region). On the other hand, if transport takes a very long time ($R_t \gg R_s$), the overall uptake will be limited by the transportation (transport-limited region). If these two processes have similar rates, both will play a critical role in determining the overall uptake (transition region). Given the gas uptake includes both mass transport and surface reaction, its limiting step can be changing as a function of γ. …"