

We thank the reviewer for the helpful additional suggestions. The error in the laminar diffusion coefficient of sulfuric acid at high altitudes has been corrected in the revised manuscript. A total of 27 additional simulations were conducted to propagate the faster laminar diffusion at high altitudes into the overall inlet transmission (Figure 5, 6, and S6), and sensitivity studies for the straight tube (Figure 7). Figures 5, 6, S6, and 7 have been revised as a result of these additional simulations. The results for simulations at low altitude (windtunnel and low altitude flight case) remain unchanged.

In the following, the reviewer comments are copied in black font, and the response is in blue font.

Major comments:

No, the authors did not fully answer my initial objections. The shortening of the paper is welcome. The wall-loss issue and relating it to a mass accommodation coefficient, m_a , is still faulty. Also, laminar diffusivity at low pressure should be properly taken into account: for the low pressure simulations they are using a diffusion coefficient that is too low by 50 % or more.

Indeed, there was an error in the laminar diffusion coefficient of sulfuric acid at high altitudes that has been corrected. This leads to a factor of two lower transmission at high altitude conditions. This change does not affect relating the wall-losses to the mass accommodation coefficient; we only have access to windtunnel data at warm temperatures and high pressure, where the correct laminar diffusivity had been used.

We respectfully disagree that our mass accommodation coefficient is faulty. In lack of any evidence by the reviewer to support his/her objections we have revised the manuscript only to make our approach in FLUENT fully transparent.

We also note, that this indirect approach yields results for α_i that are 1) consistent with our experimental observations of gas-phase H_2SO_4 loss under various operating conditions; 2) these loss measurements put tight bounds on the value of α_{SA} needed in the model in order to reproduce the observations; and 3) the inferred value of α_{SA} is found to be in excellent agreement with two previous values published in the literature (Hanson, 2005; Pöschl et al., 1998). All relevant information was provided in our replies, the revised manuscript, and the supplementary materials.

Specific comments:

- 1) The authors present the equation for diffusivity which appears to be right out of the the Fluent documentation (maybe should be referenced). But the equations used for how m_a is introduced/calculated are not shown. That may be because Fluent user guide has no section on this or it may be because the authors used a User Defined Function; if the latter it should be presented.

It is true that Fluent does not directly simulate the mass accommodation coefficient. In the revised manuscript, we are now presenting the equation we use to extend FLUENT in order to represent the mass accommodation coefficient.

The mass accommodation coefficient, α_i , is fundamentally defined as the ratio of gas molecules taken up by a surface to the total number of gas-surface collisions. Based on this definition (summarized at text book level, e.g., Finlaysson-Pitts and Pitts JR (2002)), we have constructed a ratio using the difference in mass fraction between the wall, m_{wall} , and the flow, m_{flow} , normalized by the mass fraction in the flow, to represent the variation in mass accommodation coefficient between the flow and the wall surface.

The equation we used to define our mass accommodation coefficient is as follows:

$$\alpha_i = 1 - \frac{m_{wall}}{m_{flow}} = \frac{m_{flow} - m_{wall}}{m_{flow}}$$

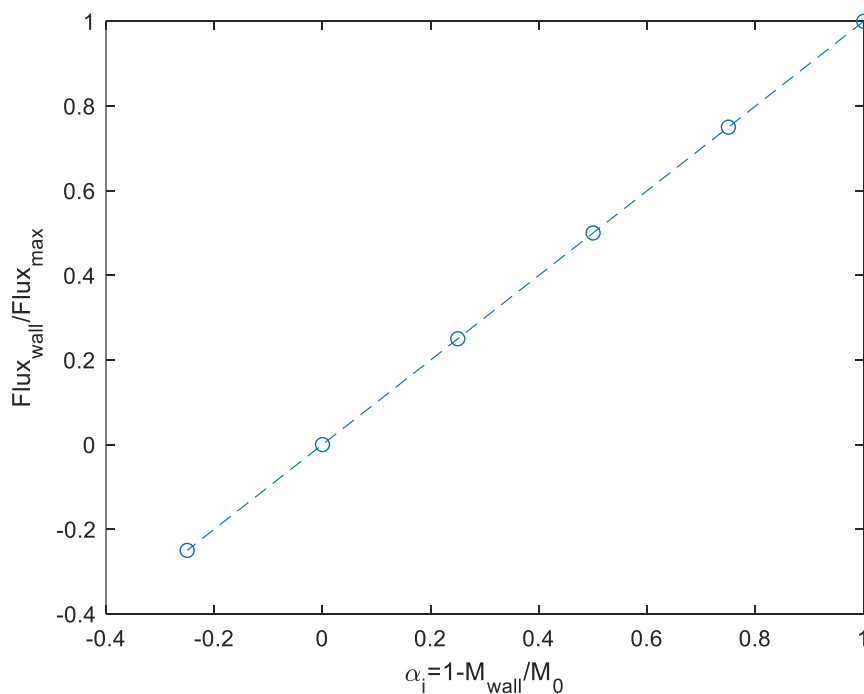
Introducing wall loss probabilities into convection-diffusion problems is tricky: I suspect this is why Fluent user guide has no information on it and it is not clear how to do it properly. The main problem here is defining m_a to be related to the mass fraction at the wall divided by the mass fraction 'in the flow'. Instead, m_a can only have meaning in these simulations if it can be related to the ratio of the mass fraction that is set at the wall to the actual mass fraction at the wall. This actual mass fraction at the wall can be taken to be the value in the cell proximal to the wall as a first attempt but then the cell thickness at the wall should be made very small, to minimize errors in gradients and extrapolating from the center of the cell to the wall. One should see the problem with setting a mass fraction at the wall constant with distance down the tube (if that is what was done rather than some UDF.) One can also see the effort needed to do a proper assessment of wall loss coefficients in CFD simulations. Currently, the simulation results without wall mass fraction set to zero (and thus unity m_a) or mass flux set to zero ($m_a=0$) are not of much use scientifically.

We agree that under unsteady-state conditions, changes in the mass fraction on the wall are unavoidable. However, unsteady-state conditions are beyond the scope of this paper.

The transmission tube is long, and it is very difficult to identify changes in the amount of species on the wall, which makes modeling the unsteady case unrealistic. Therefore, we consider each simulation to be under steady-state conditions. In addition, by simulating different mass accommodation coefficients, we are, at least, able to discuss their impact. In a real experimental environment, it is likely a combination of all these situations.

The below figure illustrates how the wall flux can be related to the mass accommodation coefficient based on the definition of α_i . As had been mentioned in the paper, we applied Fick's law of binary diffusion as our mass transport model. The mass fraction loss of a studied species represents the gas loss during transport. By directly specifying the mass fraction on the wall, we essentially simulate different mass fluxes toward the wall.

Take the following case as an example: applying Fick's law of binary diffusion to a 2D channel flow with constant velocity and diffusivity. The mass flux near the wall, as shown in the figure below, is linearly correlated with the mass accommodation coefficient α_i . If α_i is less than 1 and larger than 0, the wall flux is greater than 0, indicating that the species is being lost to the wall. If α_i equals 1, the wall flux is maximized, simulating the complete wall loss condition. If α_i equals 0, the mass fraction on the wall is the same as in the flow, as a result, the wall flux is 0, meaning no species is up taken by the wall. If α_i is less than 0, the wall flux takes a negative value, indicating that the species is released from the wall into the flow. A similar explanation can be also found in the supplementary material (FigureS5) of our previously submitted version of the paper.



- 2) Discrepancy between modeled and measurement is hard to follow and should be rewritten. At least one graph with an unequivocal comparison for a single experimental setup and model. Error bars on the measurements are necessary. The whole m_a less than unity simulation results have no meaning. Find another explanation.

We have revised the manuscript to make it easier to follow. All experimental data related to H_2SO_4 are presented with error bars, and shading in the Figures 5 and 6 illustrate error bars on the final results. Error bars were calculated using the central limit theorem and error propagation as illustrated in the Supplement.

- 3) It is not clear that the results are for the full inlet or just the straight section (see line 186, ACR version). Firstly, we stipulate to the authors' point in their initial response that the lower turbulence at low pressure is the reason the ground and low pressure simulations have similar throughputs overall (should this be one of the main findings of the paper?) Is this for both curved and straight sections? Nonetheless, loss in the 40 " tube will be affected by laminar diffusivity as, along its length, turbulent diffusivity is increasingly being confined to the center of the flow. The authors make what could be a grave mistake in not using the correct laminar diffusion coefficient and should re-run the simulations to see what difference it makes. Then this finding will have sound backing.

We have corrected an error in the laminar diffusion coefficient at high altitude; and conducted a total of 27 additional simulations to properly account for the higher laminar diffusivity at low pressure. Four figures have been updated in the revised version of the manuscript (Figure 5, 6, S6, and 7).

In the revised abstract, we now mention that at comparable Reynolds number (e.g., 2300 in revised Figure 6) the ground and low pressure simulations have similar throughputs overall.

In the revised paper, we clarify at the beginning of Section 3.5 that results of sensitivity studies are for the straight tube (this had also been mentioned in the caption of Figure 7).