

This is a short review as all aspects of the paper were not evaluated: it is too long, thus the Fair rating on Presentation. I gave it Fair on Scientific quality for reasons detailed below. I gave it Good for Significance as the loss of sticky species on inlets is important to get correct. I think with a rewrite and a careful paring down of the text along with addressing the science quality issues below, it could be a pretty good report.

Thank you for reviewing our paper and for the insightful questions. Our manuscript addresses the referee's comments as follows. We have gone through the text to pair it down as best possible. The previous sections 2.4 and 3.2 have been moved to the supplement information section. We believe that the content provided in the revised version is essential for supporting our study. It also strikes a balance to provide the reader with sufficient context about the discussions in part 1 of this paper series, without repeating information that is described in details elsewhere:
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1) Wall loss is a tricky thing to model. How is it incorporated in Fluent? Of course no loss (set species to have no flux at the surface) and diffusion limited loss (set species to have zero concentration at the surface) are conceptually easy to understand and to implement in Fluent. How does one address mass acc. coefficients other than 0 and 1 in Fluent?

As noted in Section 2.3, lines 190–201, the mass accommodation coefficient is defined as the ratio of gas molecules taken up by the surface to the total number of gas-surface collisions. In the revised paper, we have added the following text:

“ To simulate different mass accommodation coefficients, we set different species mass fraction boundary condition on the inlet walls. Using this approach, the mass accommodation coefficient is effectively set between the limits of 0 and 1, depending on the species mass fraction on the walls. The influence of varying wall mass fraction boundary conditions on the species accommodation is discussed in our previous paper (Yang et al., 2024).”

2) Related to that issue, the mass acc. results do not make sense in the laminar realm. At 298 K and using 98 for molar mass: (i) At 1 atm (or 0.85 atm? as in Colorado Springs), the diffusion limited loss rate in a 1cm ID tube is about 1.5 s⁻¹. (ii) The kinetic limit (for a mass acc. = 1) is about 2.3x10⁴ s⁻¹. What this means is that mass acc. values greater than about 0.001 are at the diffusion limit and there should be very little to no dependence on mass acc. for the throughput of the sampling tube.

While the reviewer is correct that these two rates represent one set of extremes in gas loss rates, we do not agree that these limits represent the role of accommodation coefficient on gas loss. Both the values calculated by the reviewer represent the case of unit

accommodation coefficient, applicable for very different spatial ranges (the kinetic theory for gas loss calculation is applicable if the tube ID is comparable to the mean free path – which is not the case for the 1 cm tube considered here).

As noted, at 1 atm, the diffusion limited loss rate for a 1 cm ID tube is $\sim 0.1 \text{ s}^{-1}$ (our back of the envelope calculation is off by a factor of 10 from the reviewer's), which is about 10^{-5} of the kinetic limit. Whatever the exact value of the diffusion loss rate is, it is representative for loss rate with an accommodation coefficient of 1. A lower accommodation coefficient will lower the diffusion loss from this estimate. To model the accommodation coefficient, we believe that two possible approaches could be used: proportionally scale the diffusion coefficient or tailor the boundary condition as we have done here.

3) Another related issue is the diffusion coefficient. It looks like the authors are using the pressure independent value for all pressures: it actually has units of $\text{atm}\cdot\text{cm}^2/\text{s}$. Diffusivity at altitude will be some 6 or 7 times that at sea level due to the pressure change. Yet it is colder so apply a typical $T^{1.75}$ factor. What temperature is the sampling tube at altitude?

Yes, the laminar diffusivity, D_{ij} , which describes the diffusivity between the studied species in dry air, is set as a constant. The turbulent diffusivity, however, varies with local flow conditions in the model. In turbulent flow, the contribution of laminar diffusivity to the overall gas-phase diffusion is much (orders of magnitude) smaller than the turbulent diffusivity. For comparison, the combined effect of lower pressure and temperature increases the laminar diffusivity at by about factor ~ 4 (an upper limit for most conditions during tropospheric sampling).

The original manuscript had already recognized that diffusivity significantly impacts the prediction of gas transmission efficiency, particularly within the transport tube (see Section 3.5, and the original Fig. 8, now Fig. 7). In the revised manuscript, we have added the following sentence to the discussion of Figure 7.

“The shaded area shown in Figure 7 (factor ~ 3 higher laminar diffusivity) also approximately illustrates the magnitude of the combined effect that higher laminar diffusivity at lower pressure and temperature has on the gas transmission efficiency when sampling at altitude. For most tropospheric sampling, this effect will be minor for non-sticky molecules, but it gains in relative importance for sticky molecules sampled at low pressures; which was not the focus of the current study (windtunnel experiments at ground conditions), and deserves further attention in the future for high altitude sampling.”

The temperature along the sampling tube is not really well characterized. Only the ram heating effect on turbulent diffusivity is captured in our calculations. We have clarified this in the revised manuscript with the following sentences:

“The further warming of the sample in the tube as air transfers into the instrument aboard the aircraft depends on many parameters, incl. temperature gradients towards the cabin air, flow rates, heat transfer from the tube to the gas, etc. and this is currently not well characterized and cannot be generalized; this is not easily possible to model either. We are planning to measure this gradient during an upcoming campaign to further characterize the flow conditions inside the sampling tube in-flight. “