## Final author comments

We would like to express our gratitude to the reviewer for taking the time and providing comments and a critical review to the manuscript.

Below, we provide point-to-point responses to the comments in red and document changes to the manuscript in blue.

Additional to the changes in direct context of the reviews, we have corrected multiple typographical errors in the manuscript, harmonized the panel labels of Figures 1 and 7, corrected the affiliations for one co-author, and amended the acknowledgments.

Henning Finkenzeller, on behalf of all co-authors

## Reviewer #2 comment:

This paper compares two atmospheric pressure chemical ionization geometries using a detailed model simulations. Results from ion current measurements are presented and compared to simulations of the two different geometries. The overall goal of this manuscript is interesting and the results would be useful to the community. However I find the paper inconsistent or confusing at the least.

From my understanding, the advective velocities used in the model (Figures 1 and 7) are too large for the typical operation of either case of the CIMS. This is especially true for the Eisele design. For the MION described in Wang et al. [2021] with a 22 mm flow tube, a velocity of 2 m $s^{-1}$ equates to a total flow of ~45 slpm which is on the higher end of the 20-30 slpm typically described (Wang et al. [2021] used 32 slpm for example). For the Eisele design using the 44 mm diameter flow tube described in the model, 1.7 m $s^{-1}$ equates to a total flow of ~160 slpm. Well above the 20-45 slpm typically described (Tanner and Eisele [1995] and Sipila et al. [2018] used 45 and 30 slpm respectively). Later however, in Table 1, values more consistent with previously published operating parameters are presented for both systems.

The authors state that a reaction time of 113 ms is assumed for the Eisele configuration. Using a flow of 30 slpm as stated in Table 1 and the 44 mm diameter, a flow velocity of ~30 cm $s^{-1}$ is obtained, yielding a reaction distance of only 3-4 cm? The physical length of the actual IMR cylinder is ~15 cm. The 113 ms reaction time IS greater than the calculated ~88 ms geometric reaction.

This inconsistency/confusion furthered in the topic of turbulence. The authors state that a Reynolds number, $R_e$, of ~1600 was assumed. Using 45 slpm in a 22 mm dia. flow tube, as in the MION model, yields a $R_e$ of ~2800, well into the transition zone towards turbulent flow. The 160 slpm flow in a 44 mm flow tube, as in the Eisele model, yields a $R_e$ >4000, well into the turbulent flow regime. If, however, the flows from Table 1 are used laminar conditions are maintained.

Overall, as a person familiar with both systems, I find this work inconsistant and/or confusing and hard to follow. The manuscript should be rewritten with an eye towards addressing these inconsistencies or making things more understandable. The work is interesting and will provide insight for users of either technique. The visualizations are quite useful. However, it needs more work before I would recommend publication.

Following the reasoning outlined in the “Quick reply to reviewer comment #2”, we have revised the manuscript to clarify the flow rates used in the modelling, the flow profiles, and that using laminar flow in the model is justified. Specifically, we addressed these issues in the following sections:

1. Clarification of flow rates for each inlet, Section 2.2 (Model setup)

   For the modelling of the Eisele-type inlet, 10 slpm sample, 20 slpm sheath, and 1 slpm flow to the mass spectrometer are used (Tanner and Eisele, 1995). For the MION2 inlet, 20 slpm exhaust flow (Wang et al., 2021) and 0.8 slpm flow to the mass spectrometer are used. The auxiliary reagent, purge, and reagent exhaust flow are $J_R$ = 10 smlpm, $J_{RE}$ = 50 smlpm, and $J_{RP}$ = 100 smlpm.

2. Clarification of flow profiles in both MION2 and Eisele-type, and respective Reynolds numbers (including the parameters used for their derivation), Section 3.1 and 3.2 (Results)

   MION2, Section 3.1 (Results MION2 inlet):

   Assuming an interface upstream of the MION2 inlet that creates laminar flow (Reynolds number Re ≈ 2100 (using $D = 20$ mm, $u = 1.6$ m s$^{-1}$, $\nu = 1.48 \cdot 10^{-5}$ m$^2$ s), the flow velocity profile is parabolic throughout the sample tube and IMR close up to the pinhole plate, where the flow splits to the exhaust and pinhole.

   Eisele-type, Section 3.2 (Results Eisele type inlet):

   Considering the paramount importance of the flow field throughout the inlet for the gas and ion transport, we added Figure 8 and an accompanying paragraph, which effectively communicates the modelled velocity profile within the Eisele-type inlet before and after merging the sample and the sheath flow.

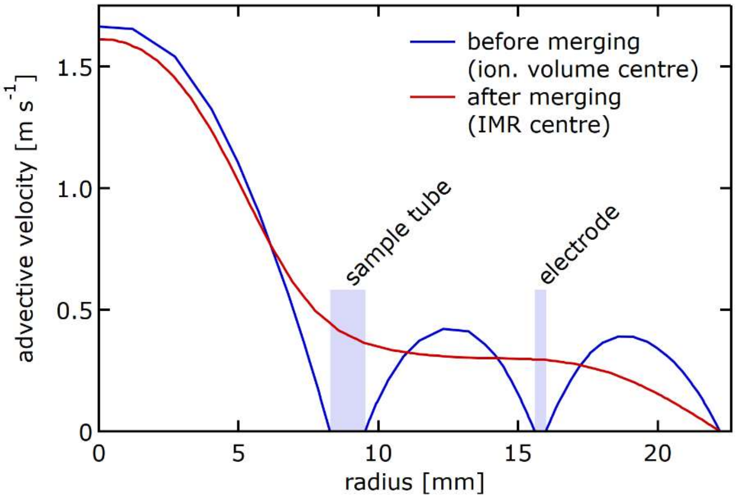

Fig. 8: Modelled velocity profile within the Eisele-type inlet before and after merging sample and sheath flow, using 10 slpm sample flow and 20 slpm sheath flow. The composite profile establishing in the IMR has a pronounced maximum in the centre and a rather flat shoulder.

> Figure 8 shows the velocity profile of the advective velocity upstream of the flow merging at the x-ray lamp plane and downstream after mixing at the IMR mid plane. Before merging, the flow profiles within the different channels are near-parabolic, as expected for fully developed laminar flows. After merging, the individual flow profiles combine to form a transition composite that maintains a near-parabolic shape in the innermost 5 mm with a pronounced maximum at the centre line and a rather flat shoulder with low velocity. The profile is the result of the relatively little interaction with the IMR surface after merging: The IMR radius (22 mm) is relatively large in comparison to the total IMR length (15 cm). Additionally, the velocity profile at the downstream end of the IMR (close to the pinhole plate) is not parabolic, either. The Reynolds number $Re \approx 1840$ (using $D = 16$ mm, $u = 1.7$ m $s^{-1}$, $\nu = 1.48 \cdot 10^{-5}$ $m^2$ s) at the downstream end of the sample tube, the location most prone to cause turbulence, supports assuming laminar flow in the modelling.

We remain convinced that there is no fundamental problem with the study and believe to have appropriately addressed the raised concerns about the clarity of presentation and consistency of the study.