The black color of the text in this document shows the reviewer's comments, while the green color shows the authors' responses, and the revised text is shown in italics.

The authors present results from a newly designed VOCUS AIM reactor. They evaluate the instrument's performance by examining its time response (focusing on sticky molecules), sensitivity, limits of detection, and selectivity. Additionally, they use dopants to mitigate the influence of humidity on the sensitivity of different molecules, with acetonitrile being the most effective dopant. The authors highlight the strength of the AIM, which can switch within 2Hz between multiple reagent ions to access compounds of varying volatility. The instrument is tested in the oxidation of α-pinene to assess the selectivity of different reagent ions and the broad access to all products of such oxidation with high sensitivity. This paper is well-written and suitable for publication. Only minor comments from my side.

We thank the reviewer for his/her careful consideration of our article. We have attached a revised version of the manuscript in which we considered all the comments raised by the reviewer.

Comments:

Line 399-403: The straightforward normalization using the recorded reagent ions measured at the detector is great. It would be helpful if the authors could clarify the differences in the AIM-VOCUS that make this possible compared to the traditional VOCUS, where such normalization can be challenging.

Unlike the Vocus AIM, the reagent ions (i.e., hydronium ions) are not detected when operating the Vocus PTR. Indeed, only the water clusters are measured with a distribution that is greatly impacted by the conditions within the reactor (i.e., temperature, DC gradient, RF amplitude). As a result, the normalization procedure cannot be compared and the direct measurement of the reagent ions with the Vocus AIM makes the normalization of the data straightforward

Line 432-436: Switching chemistries at up to 2 Hz is impressive. However, could there be any interferences from one ion chemistry to the next that were observed? Are the 2Hz switching timescales recommended to ensure no interferences? Considering that the authors have already demonstrated the impact of tailing for sticky molecules, which may take several seconds to stabilize when fluctuations occur, I wonder if such frequent switching is ideal. Discussing potential limitations in this regard would be beneficial, as it could save the community considerable time in determining the optimum operating conditions of the AIM. Additionally, it would be interesting to note how these datasets are handled from a software perspective.

The fundamental time limitation is the residence time of the ions within the AIM reactor which is 10 ms. As long as the different reagent ion precursors do not interfere with each other, their stickiness is not a critical parameter. In addition, the flow profile within the AIM reactor remains constant and no fluctuation will occur when solely turning ON/OFF the VUV lamps. Hence, the future user will need to evaluate the necessity to use high frequency based on the scope of their measurements as well as the desired sensitivity/LoD. All the data will be recorded within the same data file and will be handled in the same manner by the software.

Line 462-464: What are the expected concentrations though? The introduced a-pinene is at high concentrations and I wonder whether these compounds could be observed at such high intensity in ambient air.

Within the conditions tested in the flow tube up to ~ 4.5 ppb of AP reacted (from OH radical and O_3 oxidation) within the 70 s. Assuming a total HOM yield of 4% (Bianchi et al., 2019), the total HOM concentration should have been in an order of 180 ppt. As a result, we do expect to be able to measure HOMs under typical atmospheric conditions (~10-20 times lower compared to the flow tube experiments). Finally, the flow tube experiments were analyzed using a 1 s time resolution while ambient measurements are often analyzed using a much lower time resolution (1-15 min), which would help the detection of compounds at sub-ppt levels.

Comments on figures and tables:

Figure 1: It would be nice if the authors could indicate the pressures in the different sections of the IMR in A or B. Is the dopant flow included in the CFD calculations? I am sure the effect should be minor but still something to mention.

The pressures in the AIM reactor and the SSQ have been added in Figure 1. The CFD calculations did not consider the dopant flow, which is now mentioned in the caption in Figure 1.

Figure 3: I would recommend that the authors change the acronym of the color bar caption from "ACN" to "dopant flow".

The Figure has been modified as suggested.
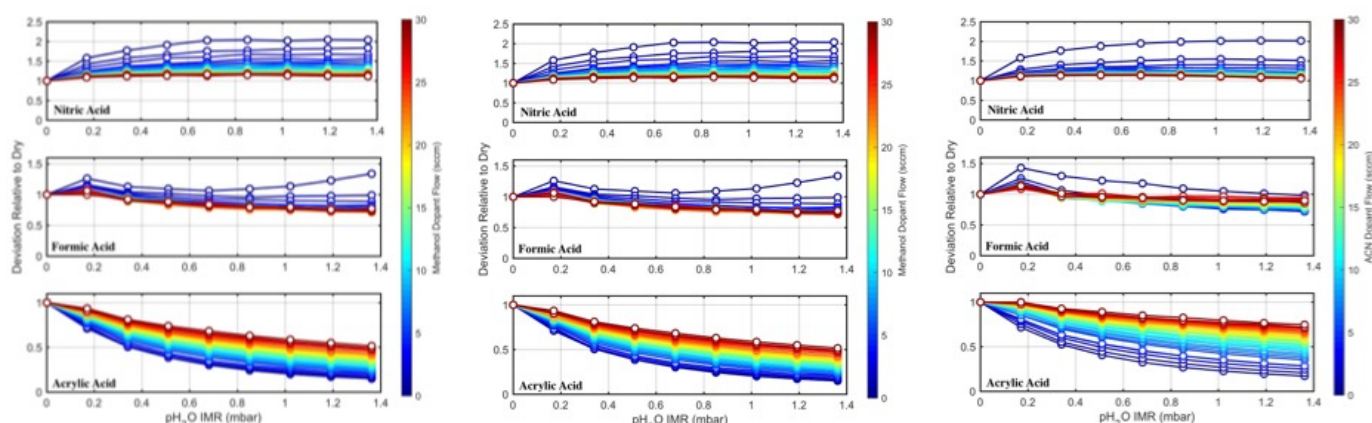
Table 1: It would be great if the authors could provide the contribution of any interfering clusters to these calibrations that could complicate the spectra. How clean are the spectra for these calibrants and what is the expected % interference of clustering? Also, more AIM reagent ion chemistries are presented including NH4+, Cl– , and NO3-. Are there separate sensitivity results for these ionization modes?

We expect that analytes for any of these ion chemistries will be detected primarily as cluster adducts, except acid compounds (e.g., sulfuric acid, polyacids,…), which are detected as deprotonated ions in negative mode such as nitrate and bromide. For detailed sensitivity results, please refer to our response to reviewer 1. However, calibrations for ion chemistries beyond those commonly used in the community were not extensively performed in this study, as they go beyond the scope of our current work.

Additionally, nitrate ion chemistry historically presents challenges in calibration, particularly because it targets very low volatile compounds that are difficult to handle and transfer quantitatively into the gas phase. We successfully calibrated the nitrate mode specifically for PFAS molecules and pesticides, achieving calibration factors ranging from 0.2 to 5 ncps/ppt, more information can be found here: https://www.tofwerk.com/pfas-detection-air-vocus-aim/

Figure 5: The yellow color is not consistent in all graphs. There is no indication of what the size is. If it is selectivity, it would be great if the authors define how it is determined in the caption.

We appreciate the reviewer's observation regarding the inconsistency of the yellow color in the graphs. We have revised the color scheme to ensure consistency across all graphs. Additionally, we have included a legend to explain the size of the points. The size of the circle is determined by the square root of the signal intensity. The caption has been revised as follows:

Lines 743-747: *"Figure 5. Mass defect plots of organic compounds measured by the Vocus AIM reactor using Ammonia, Chloride, Iodide, and Nitrate ion chemistries generated via the O3/OH initiated oxidation of α-pinene. The x-axis represents the mass-to-charge ratio of the neutral analyte, the y-axis represents the corresponding mass defect, which is the difference between their exact mass and nominal mass, and the size of the circle represents the square root of the signal intensity measured for each ion."*

The caption of Figure 6 has been also revised as follows:

Lines 758-754: *"Volatility distribution comparison for organic compounds detected by the Vocus AIM using nitrate, iodide, chloride, and ammonia ion chemistries. Ion intensity represents the cumulative signal recorded for each ion chemistry. The background colors represent the saturation concentration (C_{sat}) in the range of ultra-low volatility (ULVOCs), extremely low volatility (ELVOCs), low volatility (LVOCs), semi-volatile (SVOCs), intermediate volatility (IVOCs), and volatile organic compounds (VOCs, pink). The pie charts represent the corresponding contributions of VOC, IVOC, SVOC, LVOC, ELVOC, and ULVOC classes from the O₃/OH initiated oxidation of α-pinene.*

Typos:

Line 266: Replace "slpm" with "sccm."

This has been corrected.

Line 385: Correct to "xx%."

This has been corrected.