Response to Reviewer #2

General comment (GC):

The manuscript presents a detailed and comprehensive evaluation of the Vocus B CIMS for simultaneous measurement of volatile organic compounds (VOCs) and inorganic gaseous species. The study is generally well written and I recommend it for publication after addressing the following comments:

Response:

We thank Reviewer #2 for your recommendation and the insightful comments regarding the instrument's operation and advantages. We have addressed the concerns as follows.

Specific comment 1:

One of the main advantages of the Vocus B is its ability to measure a wide range of VOCs using different reagent ions. While this study emphasizes the synchronous measurement of VOCs and inorganic species, primarily ammonia, it would be beneficial to include mass spectra for the three reagent ion modes.

Response:

We agree that showing the mass spectra helps visualize the all-in-one capability. We have added a new figure in the Supplementary Material (see attached figure in below and Figure S2) displaying representative mass spectra for the three reagent ion modes, highlighting the primary reagent ions and typical analyte peaks. We have referenced this figure in Section 2.1.

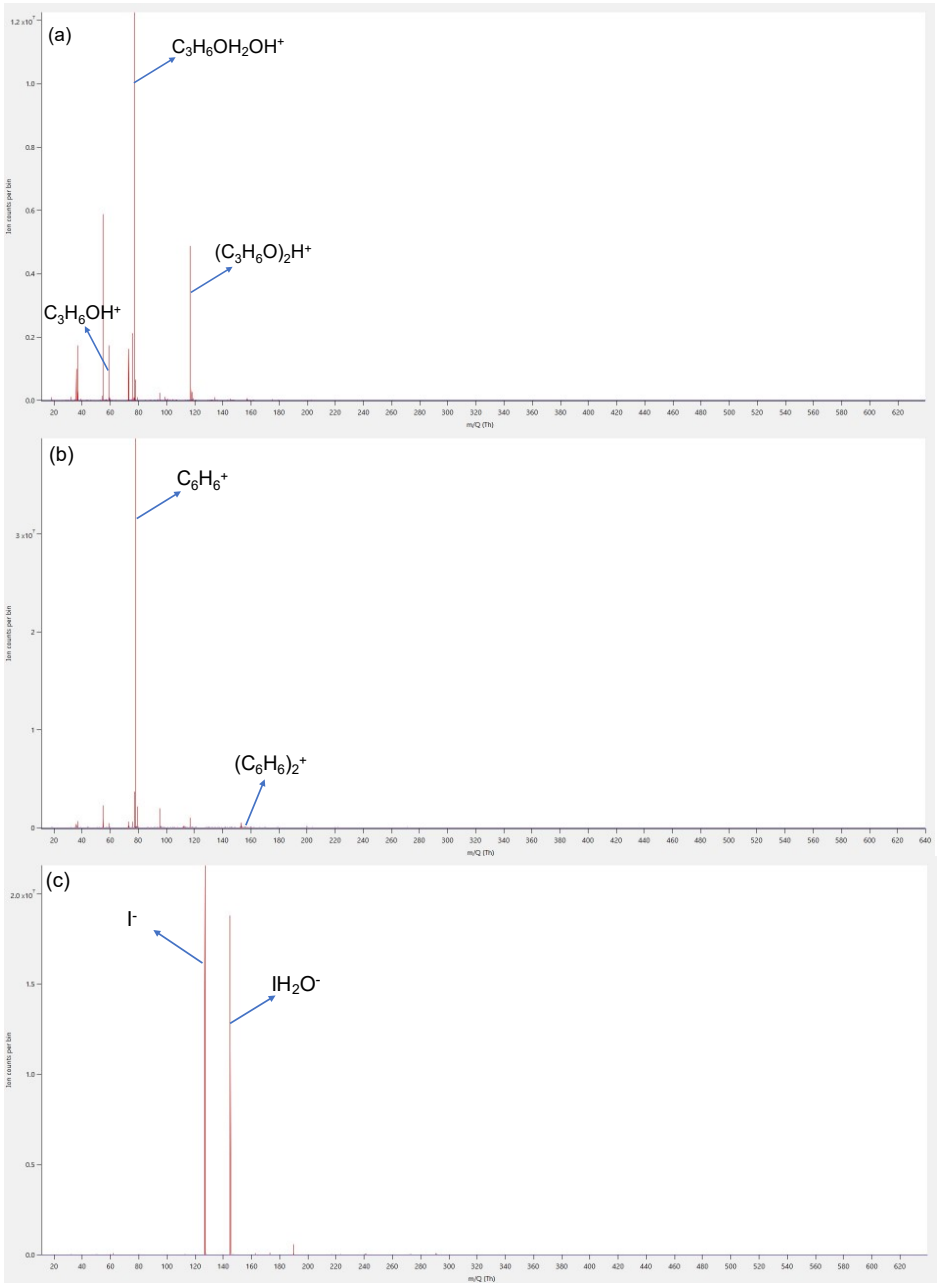

Figure S2. Representative mass spectra of three reagents, acetone (a), benzene (b) and iodine (c), in the ion mode, with the major reagent ion peaks labeled.

Specific comment 2:

A discussion on the advantages of measuring VOCs with the Vocus B compared to previous versions of the instrument would be important. Specifically, it is challenging to assess whether the Vocus B offers a clear advantage in measuring ammonia when compared to a more compact

and relatively lower-cost alternative, such as the Picarro instrument.

Response:

The primary advantage of the Vocus B is not only that it measures ammonia better than the Picarro (which is indeed a gold-standard, specialized instrument), but also measures ammonia comparably very well while simultaneously measuring hundreds of VOCs and other VICs (such as acids like HCl, HNO_3 and HF) that the Picarro cannot detect. The clear advantage is the consolidation of three separate instruments (a standard PTR for VOCs, a specialized analyzer for NH_3, and a negative-ionization (I^-) CIMS for acids) into a single platform. Compared to previous Vocus versions, the Vocus B utilizes a new AIM reactor with a conical design that minimizes wall losses, which is critical for the fast response times required for sticky molecules like amines and ammonia, as demonstrated in our mobile measurements.

We have expanded the discussion in the Introduction (near Line 65) and Conclusion to explicitly state that the main advantage is the All-in-One versatility and the reduction of instrumental footprint/complexity, rather than superior performance for a single species compared to a specialized analyzer.

Revised Introduction: "...To overcome the need for such analytical compromises, here we introduce the Vocus B Chemical Ionization Time-of-Flight Mass Spectrometer (CI-TOF-MS)... **Unlike specialized analyzers (e.g., CRDS) that offer benchmark precision but are limited to single species, the Vocus B is engineered to provide a unified "all-in-one" solution.** This platform consolidates the capabilities of multiple separate analyzers by measuring both VOCs and VICs simultaneously with sub-second switching between optimized reagent ions. This significantly reduces the instrumental footprint and operational complexity while preserving the temporal correlation between chemically diverse species."

Revised Text:

"...It effectively addresses the long-standing analytical challenge of concurrently measuring VOCs and VICs. **While specialized techniques like CRDS remain the gold standard for**

ultimate precision of individual species like NH_3, the Vocus B offers a crucial advantage in versatility. By successfully integrating the measurement of organic and inorganic compounds into a single platform, it minimizes the logistical burden and synchronization errors associated with deploying multiple separate instruments. The instrument's success across three distinct applications..."

Specific comment 3:

Additionally, a detail that seems to be missing is the cycling process between the three ionization modes. How long does the instrument operate in each mode? Could the authors clarify the time allocated to each mode in the cycling process?

Response:

We apologize for the omission. The instrument is capable of switching modes in under 500 ms. In the field campaigns presented here, we typically employed a cycle where the instrument dwelt in each mode for approximately 500 ms to 1 second, resulting in a total cycle time (and data resolution) of approximately 2 seconds (0.5 Hz) for the full suite of measurements.

We have added specific details about the duty cycle and dwell times used in our experiments to Section 2.1 (Materials and Methods).

Specific comment 4:

Furthermore, the calibration of VOCs was conducted using the benzene reagent ion mode. It would be helpful to know if the authors have also conducted calibration for VOCs using the other two reagent ion modes (Figure 1). This would allow for a comparison of results across the different modes and provide insight into the consistency of measurements.

Response:

While some VOCs can indeed be detected in positive Acetone and negative Iodide modes, the

Benzene mode (Charge Transfer) is chemically optimized for the broad detection of non-polar and aromatic VOCs with the highest sensitivity. Therefore, we focused our calibration and quantification of VOCs on the Benzene mode. Calibrating all VOCs in all modes is not standard practice for this instrument, as we prioritize the data from the mode best suited for each compound class (Benzene for VOCs, Acetone for Amines/NH3, Iodide for Acids/Oxygenates).

Specific comment 5:

The term "volatile inorganic compounds" may not be entirely appropriate in this context. I suggest using "inorganic gaseous species" or another more specific term, especially since the focus in this study is primarily on ammonia.

Response:

We appreciate the reviewer's suggestion regarding terminology. Respectfully, we have decided to retain the term "Volatile Inorganic Compounds (VICs)" in this manuscript. Our reasoning is twofold:

First, the term "VICs" provides a direct and logical parallel to "VOCs" (Volatile Organic Compounds), which is the other main subject of this study. Using "VOCs and VICs" highlights that both groups are gas-phase constituents with comparable volatility that are being measured simultaneously by the same mass spectrometric technique.

Second, the abbreviation "VICs" significantly improves the flow and conciseness of the text, particularly when discussing the co-existence and interaction of organics and inorganics repeatedly throughout the manuscript.

We believe that in the context of this "All-in-One" instrument paper, this terminology effectively conveys the intended message.