Response to reviewer #1:

We thank the reviewer for the review and suggestions. The reviewers' comments are written in blue and our point-by-point responses are in green.

The paper is well written and covers a topic of great interest for updating the ion sources on CIMS. With the inclusion of the brief tests of the cations the paper is even more pertinent. Recommend publishing with the following issues to be addressed.

Line 29: isobars should be isotopes

Our high-resolution time of flight mass spectrometer can resolve both isotopes and isobars, and the isobars (molecules with the same nominal mass, but with different formulae) cannot be resolved with lower resolution instruments. We prefer to use isobars to accurately represent the capability of this instrument.

Line 36: Unclear sentence 'Relative to nitrate...' Nitrate CIMS cannot measure amines and highly oxygenated organics

Nitrate was indeed used to detect highly oxygenated organics and amines, specifically dimethylamine, as shown in the paper we cited in the original text: Simon et al., 2016.

Line 88: units of pressure in formula is Pa while the rest of the paper is mbar. Should change to mbar

Thank you for pointing this out, and we have changed the units to [mbar]

Line 178 & 229: Is 18sccm of water vapor really added or 18sccm of saturated N2 added?

There is no N2 added through the water reservoir, and 18 sccm is entirely water vapor. The low operating pressure allows the addition of pure water vapor from the headspace of a water reservoir at room temperature (water vapor pressure at 22 degC is approx 26 mbar). We use the same delivery system as standard H3O+ instruments with a flow controlled by a low delta P MFC that comes with the Vocus PTR-TOF.

Line 139ff now reads: "A separate inlet port for humidification supplies pure water vapor from the headspace of a liquid water reservoir to the sample gas, typically between 10 and 20 sccm, using the low-Δp mass flow controller (BRONKHORST HIGH-TECH B.V., the Netherlands) originally shipped with the Vocus PTR-TOF."

Line 178 now reads: "...the drift tube pressure was 9 mbar, with 18 sccm of pure water vapor added." (Added the word pure);

Line 229 now reads: "...while 18 sccm of pure water vapor was directly added to the drift tube."

Line 315: Depending on the C3H6 to NH3 concentration if both are added to the tube illuminated by the VUV lamp, the reaction scheme could be:

1) C3H6+ + C3H6 >> C3H5 + C3H7+

2) C3H7+ + NH3 >> C3H6 + NH4+

As the PA of C3H5 (736) is less than that of C3H6 (751).

That is very true. We updated the mechanism. Thank you!

<u>Scientific significance</u>: The paper is if great significance now with the continued development of a replacement for the radioactive ion sources used previously. Not only are they safer, but even generate higher sensitivities.

<u>Scientific quality</u>: Experiments in the paper are well done with a very good description of the instrument used.

Presentation quality: Well done and no recommended changes.