Anonymous Referee #1
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Some points which should be addressed are listed in the following:

We thank the reviewer for their careful reading of the manuscript and have included responses to the reviewer's comments below and indicated where changes were made to the text.

Page 389, line 4 (and also line 25): *Why are the standard ^{210}Po not suitable for the operation at lower pressures and flow rates? It would be nice if this could be explained in one or two sentences.*

Commonly used ^{210}Po ion sources (such as the NRD models P-2021 and P-2031) are end-of-line ionizers designed for use in high pressure sprayers to neutralize static charges on surfaces in industrial processes. The sources have small diameters and small diameter to length ratios and rely on high flow rates to propel the ions out of the source before ion recombination or loss to the walls occurs. The number of ions produced per alpha particle is a linear function of both the path length the alpha particle travels in the source volume and the pressure within the source, so the combination of small diameter with low source pressure leads to a small numbers of ions produced. The higher diffusion rates at low pressure and longer residence time with low flow rates leads to greater loss to the walls. The sources also typically have screens in at the inlet/outlet to prevent damage to the radioactive foils, which provides additional surface on which ions can be lost.

The sentence "At low pressures, fewer ions are produced per α particle, and at low flow rates, the loss of ions to surfaces and recombination before they can exit the source is increased" has been added to the text.

Page 392, line 13: *"flows (0-20 sccm each)": the flows indicated in Fig. 1 are somewhat different*

The flow controllers were 20 sccm full scale, but full flows were not used for the higher two H_2 standards. The text has been changed to read "a series of stepped flows (≤ 20 sccm each) from three H_2 standards"

Page 392, line 27: *How is the [H_2O] in the zero air known ([H_2O] is used to denote the water vapor concentration)? What is the exact water vapor concentration in the zero air? Throughout the manuscript, mostly a range between 0.5 and 0.8 ppmv is stated (e.g. Fig. 1; page 396, line 11; page 398, line 7). However, at other places (e.g. Table 2; Fig. 5, lowest calibration point) it seems that this range is much smaller and that the zero air water vapor content is much better defined as the range from 0.5 to 0.8 ppmv suggests. It should be made clear whether the range refers to the variation in [H_2O] for different zero air gas bottles, or if it is due to the uncertainty of the [H_2O] in one zero air gas bottle.*

The H_2O content of the zero air was determined for each flight cylinder using the MBW frost point hygrometer. Mixing ratios were determined before and after each flight to insure nothing changed. This is stated section 3.2.1 and has been added to the section describing the MBW (3.1.3). The text has been changed to explicitly state that the H_2O in individual zero air cylinders ranged from 0.5 to 0.8 ppm.

Page 396, section 3.1.3: *A chilled mirror hygrometer (MBW) is used as the reference instrument for the calibration. Therefore, the question is why the MBW is then not used in combination with the calibration system during the flights. As it is demonstrated later (from the data shown in Fig. 11 and Fig. 12) it becomes clear that the CIMS instrument has some important advantages over the MBW in terms of precision and time response. Therefore, it might be useful to point the reader already to these benefits in section 3.1.3 to avoid confusion.*

The MBW is a commercial bench-top instrument not designed for operation in harsh conditions (such as the low T and P environment of the WB-57 pallet bay), and the time constant of the instrument, especially at low H_2O concentrations, limits its use for dynamic measurements.

A sentence has been added to section 3.1.3 stating "While the MBW has both high accuracy and precision, it operates best as a steady-state instrument and has a slow time response at low water vapor concentrations that limits its direct use for dynamic measurements such as from aircraft in the UT/LS."

Page 399, line 13: *There seems to be a typo in "in quantities to that produced H_2O mixing ratio".*

Text changed to read "to produce H_2O mixing ratio increases".

Page 401, line 27: *It should read "descended back to 13.9 km".*

Text corrected as suggested.

Page 403, line 9: *It is not clear what the word "instrument" refers to here. Do the authors mean the vacuum chamber of the mass spectrometer?*

The sentence has been removed.

Page 418, Fig. 8: *Two of the steps in region IV show rather large spikes in the H_3O^+ signal. Where do these come from and can it be ruled out that they influence the calibration curve?*

Text added to the figure caption discussing the cause of the spikes and their lack of effect on the calculated averages for the calibration steps.

Table 2: *It would be good to state also the overall uncertainty (systematic and statistical error) in the measurement.*

Line added to Table 2 stating overall measurement uncertainty.

Fig.2: *Indicating the direction of flight would be beneficial.*

Flight direction indicator added to Fig. 2.

General comment: The unit "ppmv" should be used consistently (as in Fig. 5), this is not always the case (e.g. "ppm" in Fig.6).

Text changed to consistently use "ppm", which is explicitly defined as 10^{-6} mol/mol at first usage.