Dear reviewer #1,

thank you for reviewing our article on trace gas measurements in the UTLS using the airborne chemical ionization mass spectrometer AIMS. We appreciate the corrections and new suggestions to the manuscript. Particularly the background measurements during flight and the inflight calibration were added and discussed in more detail in the script. We also address the response time of the inlet to HNO_3 transmission. We hope, that the paper is now compelling and conclusive.

Below we try to address the comments point by point. Our answers are written in italic/cursive letters.

Kind regards,
Tina Jurkat

Comments and answers to Referee #1

The paper describes an airborne CIMS instrument for the measurement of SO2, HNO3, HCl, and HONO. Instrument characterization, calibration, and some initial measurements are described as well. A novel, discharge type ion source is described. The paper is quite long considering that the CIMS technique using SF5- ions has been around for a long time. maybe the authors could consider shortening the manuscript by eliminating some of the background discussion of ion chemistry and description of theoretical sensitivity as it does not apply anyway in the end.
I have no major concerns with the manuscript. There are a few qualitative statements that could be backed up better, such as:

We have shortened Section 5.1 on "Retrieval of trace gas concentrations for AIMS-TG" and included instead a figure and paragraph on the inflight calibration to address the sensitivity derived in flight.

p. 13587 / l. 25 what was the characteristic time scale of this exponential decrease?
The background of HNO_3 and HCl over the whole flight was best described with the following fit function

$$y = y0 + Ae^{-(x-x0)/t}$$

With characteristic time scales o t=28-50 min. Thus the background was generally at a constant level after one hour depending on the humidity of the local environment where HALO was stationed. We propose to demonstrate the effect of the background decrease with an additional plot in the supplement material (Figure 2) showing the behavior of the background measurements for the flight on 11 September 2012. Additionally we show 5 min background measurement sequence for HNO_3, HCl and SO_2 in Figure 6.

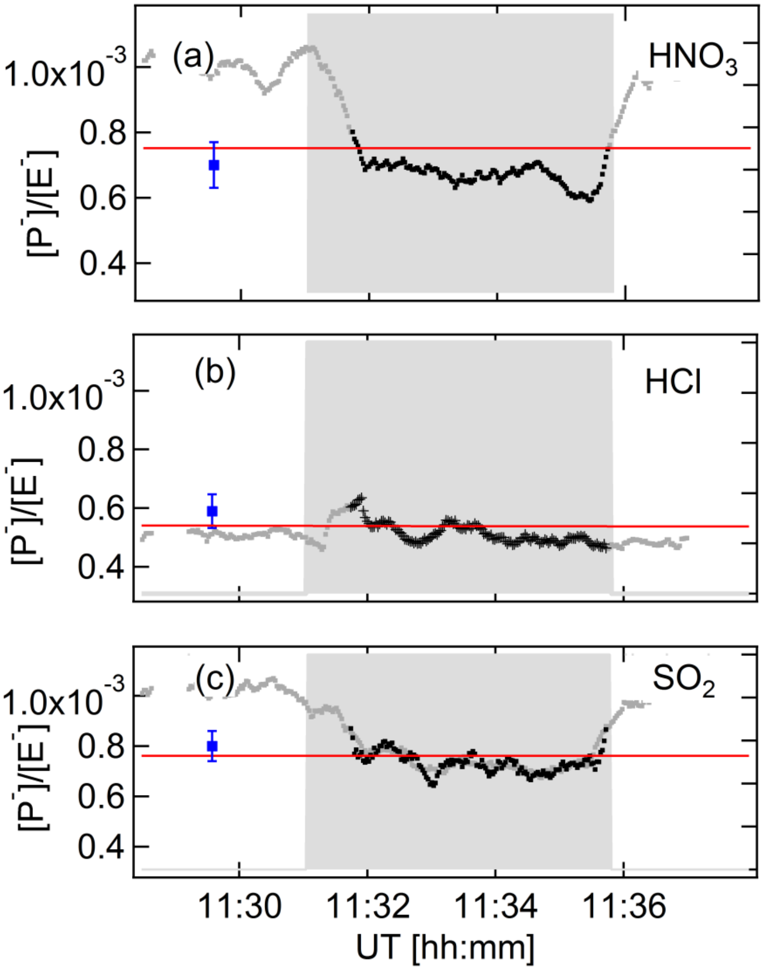

Figure 6 Ion ratios during background measurements for (a) HNO_3,(b) HCl and (c) SO_2 in tropospheric air during the flight on 11 September 2012 at 11:31 UT. Grey shaded areas show sequences where synthetic air was introduced. The red curves represent the values applied to correct for the instrumental background during atmospheric measurements. Except for the first hour of the flight, the background can be described by a constant value of 0.75e-3 for HNO_3, 0.54e-3 for HCl and 0.8e-3 for SO_2. For comparison, the ion ratio averages inferred during laboratory measurements are given (blue squares). Generally, flight and ground based measurements agree within the given variability of the laboratory measurements. Background measurements in stratospheric air are demonstrated in Figure 5.

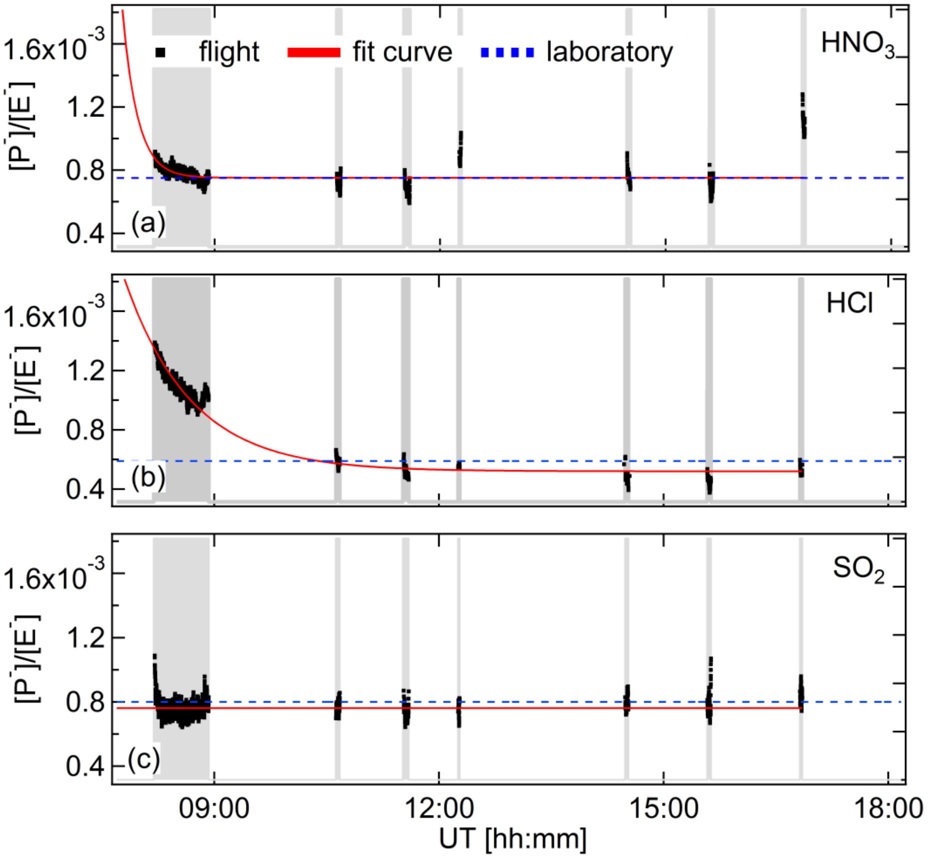

Figure 2 (Supplement Material) Ion ratios during background measurements for HNO_3, HCl and SO_2 during the flight on 11 September 2012 .Grey shaded are time sequences where synthetic air was introduced and a stable ion ratio was observed. The red curves represent the fit curves applied to correct for the instrumental background during atmospheric measurements. Generally during the first hour, the background follows an exponential fit. For the rest of the flight, the background can be described by a constant value. For SO_2, a constant background value of 0.008 was used for the entire flight. For comparison, the ion ratio averages measured during laboratory measurements are given (blue dashed line). Generally, flight and ground based measurements agree well.

p. 13591 / l. 16 - how is the expected HONO mixing ratio calculated? This should be difficult as nothing is known about the history of the air mass during convective transport, especially with respect to j-values and heterogeneous processes.

The initial HONO mixing ratio is calculated using an e-folding life time of 10 min at day time. This is in fact a rough estimate and only valid in the morning hours in low latitudes in cloud-free air. Since we do not assume transport from the boundary layer but rather in-situ production, heterogeneous processes should not play a big role. Light attenuation in optically thick clouds may only enhance the HONO mixing ratio. Therefore we term it a lower limit of the initial HONO mixing ratio.

Another concern is the slow time response of the calibration. This is a problem for in flight measurements, as the characteristic time should be mostly independent of

absolute mixing ratio, contrary to what is stated on page 13591 / line 16 and below. I think this issue deserves more insight, some of which could come from the comparison with the NOy instrument shown in Figure 7. Unfortunately the small plot shows three hours of data so the details of the relative time responses of the two instruments cannot be seen. The analysis of fine time scale data from the two measurements along with laboratory tests eliminating one or more of the inlet components should shed some light on this. A 8-minute characteristic time for HNO3 is really not acceptable for aircraft measurements.

These concerns should be addressed before the manuscript is published.

We agree that the discussion of the time response is important and cannot be addressed with laboratory calibrations. Comparison to other instruments on HALO like AENEAS only yields a relative characteristic response time. Here we use the t_{90} *times which is the time until 90% of the stable signal has been reached and call it response time.*

We therefore suggest to add another plot with two inflight calibrations as a comparison to the ground calibration. In Figure 5 the time series of the ratio of product to reagent ion [P-]/[E-] for HCl, HNO_3 *and* SO_2 *measured in flight is shown for two campaigns TACTS/ESMVal in 2012 and POLSTRACC in 2015/2016. A 12 min calibration sequence performed in flight during TACTS/ESMVal is shown in Figure 5a. 5.8 ppbv of* HNO_3 *and 2.6 ppbv HCl was added to the total sample flow. The two gases were successively added to investigate the effects of the presence of other trace gases on the ion ratio.* SO_2 *was continuously added. The short increase of* SO_2 *after addition of* HNO_3 *is due to unstable flows. In Figure 5b,a calibration performed during POLSTRACC, we added 3.5 ppbv* HNO_3 *and 2.65 ppbv HCl simultaneously to the total sample flow.* SO_2 *was not online calibrated during the mission. Both calibrations were followed by a background measurement. Due to an improvement of the calibration set up (shorter lines, additional valves, continuous operation of the permeation ovens) during the second campaign, a clearer picture of the inlet line's response times evolves: HCl stabilizes after 20s,* HNO_3 *after 30s. Both campaigns had the same inlet flow and lengths. While inlet response times* t_{90} *of 20s for background measurements are the same, the calibration response time was improved for the POLSTRACC campaign. Absolute values of the calibration signal disagree between the two campaigns due to a different ion source design.*

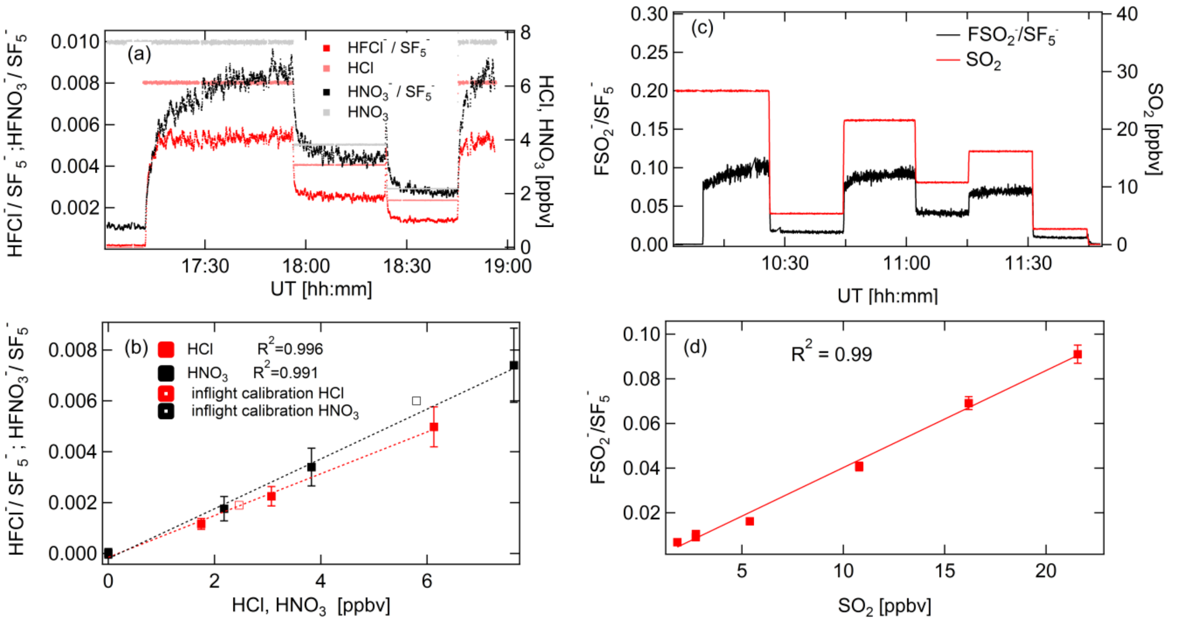

Figure 4 Laboratory calibrations of HNO_3, HCl and SO_2. (a) Time series of product to reagent ion ratios for HCl (red) and HNO_3 (black) are shown. While HCl reaches a plateau within a few minutes, HNO_3 needs more time for passivation. Once the sampling line is passivated, the plateau for a given concentration is reached faster for both trace gases (here after 18:45 h). Additionally the concentrations of the added calibration gas are shown in light red and grey. For comparison, inflight calibrations of HNO_3 and HCl are included, represented by the empty square. (b) Calibration curves for HCl (red) and HNO_3 (black). The calibration is linear up to high concentrations of 6 ppbv which can occasionally be found in the lower stratosphere. The error bars denote the precision of a single measurement. Figure 4 (c) and (d) is analogue to (a) and (b). In contrast to HCl and HNO_3, SO_2 is calibrated for a wider range of concentrations to account for high mixing ratios in volcanic or aircraft plumes.

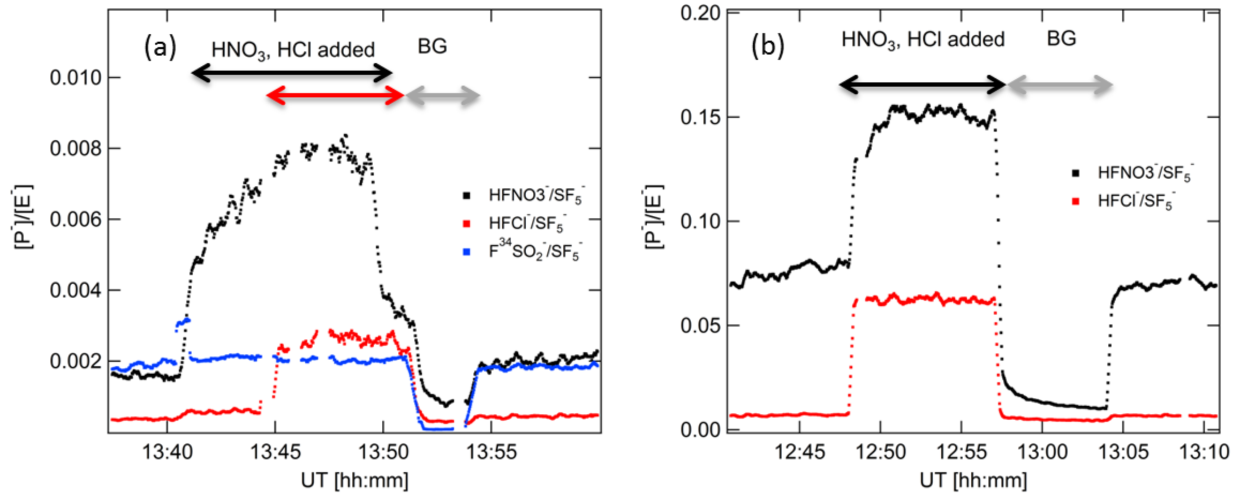

Figure 5 (a) Inflight calibrations of HCl, HNO_3 and SO_2 during TACTS/ESMVal. The time series of the ratio of product to reagent ion $[P^-]/[E^-]$ for a 12 minute calibration sequence is shown. 5.8 ppbv HNO_3 was added to the total sample flow for approximately 10 min and stabilizes after 4 min. After 3 min, 2.6 ppbv HCl was added for 8 min and showed a stable signal after 20 s. 2.9 ppbv SO_2 was added continuously. A background measurement (BG) was performed shortly after the calibration sequence with t_{90} of 15 s for SO_2 and HCl, and 20 s for HNO_3, respectively. (b) Inflight calibration of HCl and HNO_3 during POLSTRACC. 2.6 ppbv HCl and 3.5 ppbv HNO_3 were added simultaneously for 8 min. t_{90} for the calibration signal is 15 s for HCl and 20 s for HNO_3. Background measurements were performed shortly after the calibration with t_{90} of 15 to 20 s. Improvements of the calibration system between the two campaigns significantly enhance the speed of the calibration measurements. While inlet response times t_{90} for background measurements are the same, the calibration response time was improved.

Technical comments and minor issues:
Abstract line 15 – what is meant by "a characteristic ionization scheme?"
*We use the word for an overall description of the reaction and fragmentation pathways. This wording may not be of general understanding thus we replace the phrase with "*For AIMS-TG, a custom-made gas discharge ion source has been developed for generation of reagent ions that selectively react with HCl, HNO_3, SO_2 and HONO*".*

Abstract l. 19 – write "low 10s of ppt range"
Abstract l. 22 – replace "expemplarily" with "for example"
Throughout manuscript – use "l" instead of "L" for units of liter
Page 13569 l. 13 replace "great" with "large"
13572 l. 15 and throughout rest of manuscript - replace "casted" with "cast"
We made the suggested changes.

13575 / 5 – was this technique employed here?

Yes, it was employed. When choosing the potential to the ion source needle tip we adjust the voltage such that the lowest background on the specific masses evolves at the highest reagent ion count rate. In addition the orientation of the needle with respect to the pinhole determines the stability of the discharge and therefore the background counts. We suggest demonstrating the effect of the ion source voltage on the background signal in an additional plot as a supplement material (Figure 1).

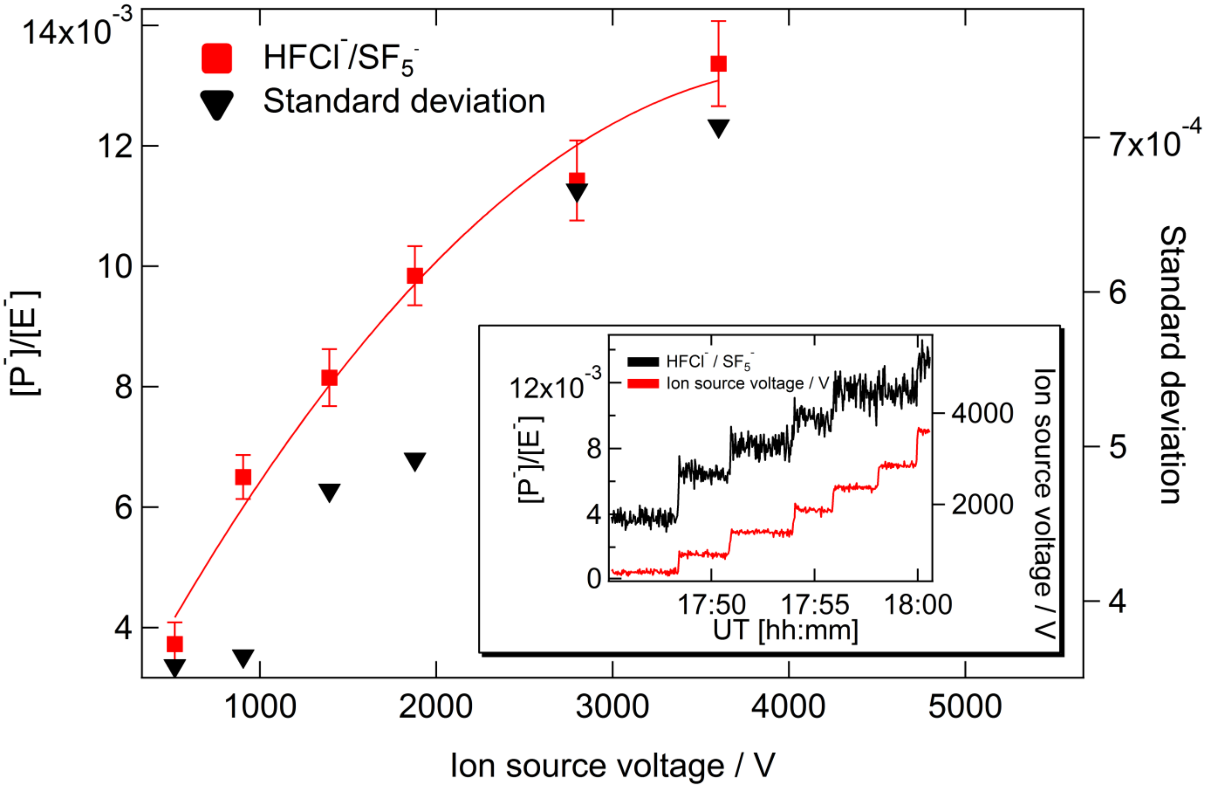

Figure 1 The ratio of product to reagent ions [P$^-$]/[E$^-$] (here HFCl$^-$/SF$_5^-$) during background measurements is shown for different ion source voltages. The inset shows the timeline of the signal, while the larger plot shows the direct correlation of the average values of [P-]/[E-] and the standard deviation of the background signal for different ion source voltages. With increasing voltage both background level and standard deviation of the background increase. Generally voltages between 400 and 1400V are used for the AIMS ion source.

13576 / 5 – replace "in" with "of"
13576 / 10 – replace "analogically" with "in the same way"
We made the suggested changes.

13580 / 25 – how many measurements of the permeation rate were made during these 3 years? Just two? Is a linear decrease assumed?

All together 5 calibrations were done, mainly centered around the campaigns, two in 2009 and three in 2012. Yes, a linear decrease was assumed.

13581 / section 4.2 – this paragraph describes issues with calibration and sensitivity but the statements are vague and general, the authors should be able to provide more precise results.
We have included a few more quantitative measures to the section. Particularly for the calibration and background measurements we have added Figure 5 and Figure 6.

13585 / 18 – is that true for thunderstorm outflow (as shown in figure 5)
In general, it may not be true for every thunderstorm. We have certain characteristic reactions that occur in the presence of water vapor. In cases, when we expect a change in calibration factor we see an increase in mass 39 (FHF^-*) which comes from a reaction of *SF_5*- with *H_2O*. For *$H_2O>100$* ppmv, the calibration factor changes. This was not the case for the thunderstorm but for the dive. Here, the data below 8 km were discarded due to water vapor intereferences.*

13587 / 25 what was the characteristic time scale of this exponential decrease?
See answers above. Answer: The background was best described with the following fit function

$$y = y0 + Ae^{-(x-x0)/t}$$

With characteristic time scales of t=28-50 min. Thus the background was generally at the a constant level after one hour depending on the humidity of the airport where HALO was stationed. We suggest to include Figure 6 in the main script and Figure 2 in the supplement material to show the behavior of the background on short and long time scales.

13588 / 19 – I do not understand this. How does the effect of flushing the lines on the ground extend past the characteristic time scale of the system, e.g., 500s for HNO3??
We agree the passivation on the inlet walls may easily be washed off in the humid air at lower altitudes. We therefore suggest to rephrase the sentence and focus more on this effect for the calibration lines. They are generally less affected by humidity changes and therefore keep the passivation.

*"Passivation is achieved by flushing the calibration line on the ground before take-off with the *HNO_3* and HCl which reduces the response time of the calibration. "*

13590 / 20 – figure 5 does not sow a flight pattern. Rephrase sentence "Results from a typical flight: : :"
13593 / 12 replace "stabely" with "stably"
We made the suggested changes.