Dear reviewer #3,

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 try to address the response time of the inlet and calibration lines with respect to trace gas transmission. We added the flows to Figure 1 and inflight detection limits in Table 1.

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

Kind regards,
Tina Jurkat

Comments and answers to Referee #3

The manuscript "The airborne mass spectrometer AIMS – Part2: Measurements of trace gases with stratospheric or tropospheric origin in the UTLS" by Jurkat et al. describes a new instrument that performs sensitive in-situ measurements of important trace gases in regions of the atmosphere that are under-sampled and difficult to access. These new measurements will provide valuable insights into UTLS processes.
The instrument capabilities are clearly illustrated with results from an instrument flying in the upper troposphere. Considerable effort has been invested in calibrations and characterizing measurement sensitivity. The in-flight calibrations and background determinations are critical diagnostics for achieving accurate measurements, and the in-depth descriptions of these efforts represents a substantial advancement. Though the instrument clearly works and provides unique and useful results, there are several aspects of the paper that need to be clarified for these findings to be useful to other researchers. In particular, the instrument duty cycle, time response, background, and precision needs to be better quantified.

The instrument time response is confusing, and this further confuses the interpretation of the results. I recommend making a single section that details the time response for each compound. The abstract states 1 s time resolution, Table 1 uses 1 s for some species and 20 s for others, and section 6.2 characteristic times are listed as 500 s for HNO3 and 150 s for HCl. The timeseries in figures 5 and 7 do not state the time averaging. Is it 1s, 20s, 500s? I don't understand the precisions listed in Table 1 and I don't think the precision should be represented as a percentage. At low mixing ratios, counting statistics should cause a large fractional imprecision that diminishes with larger mixing ratios. HNO3 is listed with a 20% precision, but the data shown in Figure 7 look far better than that. On the other hand, if the inlet integrates for 500s, then determining a precision with a 20 s average isn't meaningful. Table 1 should also include a column that lists time response, especially if it varies between compounds.

We try to clarify the instrument's time response with two additional figures and two new columns with inflight detection limit and response time (t_{90}) in Table 1.We define the response time t_{90} the time until the calibration signal or background signal has reached 90% of the expected stable value. In order to separate the response time of the inlet from the response time of the calibration system we show two inflight calibration plots. For the second calibration plot, the calibration system was significantly improve by modification of the length and passivation of the calibration lines, the temperature stability of the oven, and one additional valve in front of the *calibration line towards the inlet line.*

Figure 5 shows the inflight calibration from two campaigns, TACTS/ESMVal in 2012 and POLSTRACC in 2015/2016 with the ion ratios of HCl, HNO_3 and SO_2. In Panel (a), an inflight calibration of HNO_3 reaches 90% of the maximum signal after 4 min. HCl and SO_2 show shorter response times of generally 20 s or less. Background measurements have been performed shortly after the calibration. 90% of the background values are reached after t_{90}=15 s for HCl and SO_2 and t_{90}=20 s for HNO3, respectively. During TACTS/ESMVal SO_2 was calibrated continuously. The short enhancement of the SO_2 signal after addition of HNO_3 can be explained by unstable flows. The long response time during calibration (inflight and in the laboratory) during this campaign is mostly due to the small flow through the calibration lines and their passivation and in general not representative for the response time of the inlet. Figure 5b shows a calibration sequence obtained during POLSTRACC where we added 3.5 ppbv HNO_3 and 2.6 ppbv HCl simultaneously to the total sample flow. 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 response times evolves: HCl stabilizes after 20 s, HNO_3 after 30 s (t_{90}=15s and 20s). Both campaigns had the same inlet flow and lengths. While inlet response times t_{90} of 20 s for background measurements are the same, the calibration response time was improved for the POLSTRACC campaign.

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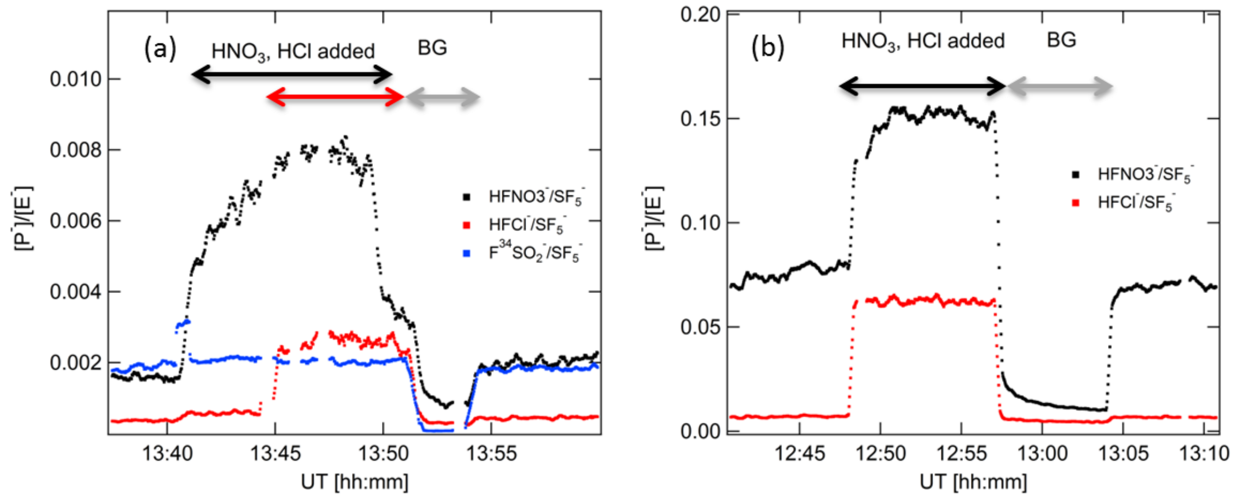

Figure 5 (a) Inflight calibrations of HCl, HNO_3 and SO_2 during TACTS/ESMVal. The time series of the ratio of product to reagent ion $[P^-]/[E^-]$ for a 12 minute calibration sequence is shown. 5.8 ppbv HNO_3 was added to the total sample flow for approximately 10 min and stabilizes after 4 min. After 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.

Additionally, the correlation of HNO_3 and HCl, with a correlation coefficient of 0.979 as well as the correlation coefficient between HNO_3 and NOy of R^2=0.98 indicate that the response time of HNO_3 is comparable to that of HCl and to NOy measured by AENEAS. Thus, given HCl *response time of 15 to 20 s, we argue that HNO_3 has a similar response time. Otherwise the correlation wouldn't be as compact.*

The background levels are determined several ways and they are discussed qualitatively, but they are never quantified. What are the background levels? Are they the same for all compounds? What is the magnitude and time constant for the exponential decrease in background (pg 13587, line24)?
Is this decrease the same for all compounds?
Is it the backgrounds that establish the detection limits in Table 1? Given the instrument's high sensitivity, the detection limits could be much better in principle. It would be helpful to understand what determines the detection limit. Nylon wool is used to determine instrument background sometimes. This should remove HNO3, but will it remove HONO SO2 and HCl from an airstream? Has this been tested? It isn't clear how the background determinations are used in the data analysis. How is the background subtracted? Do you interpolate between all the backgrounds?
To clarify how the background was retrieved we propose to add figure 6 to the script showing a 5-min background measurement and Figure 2 to the supplement material showing the long term evolution of the background.
The long term 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 of 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. Figure 2 shows the behavior of the background measurements for the flight on 11 September 2012.

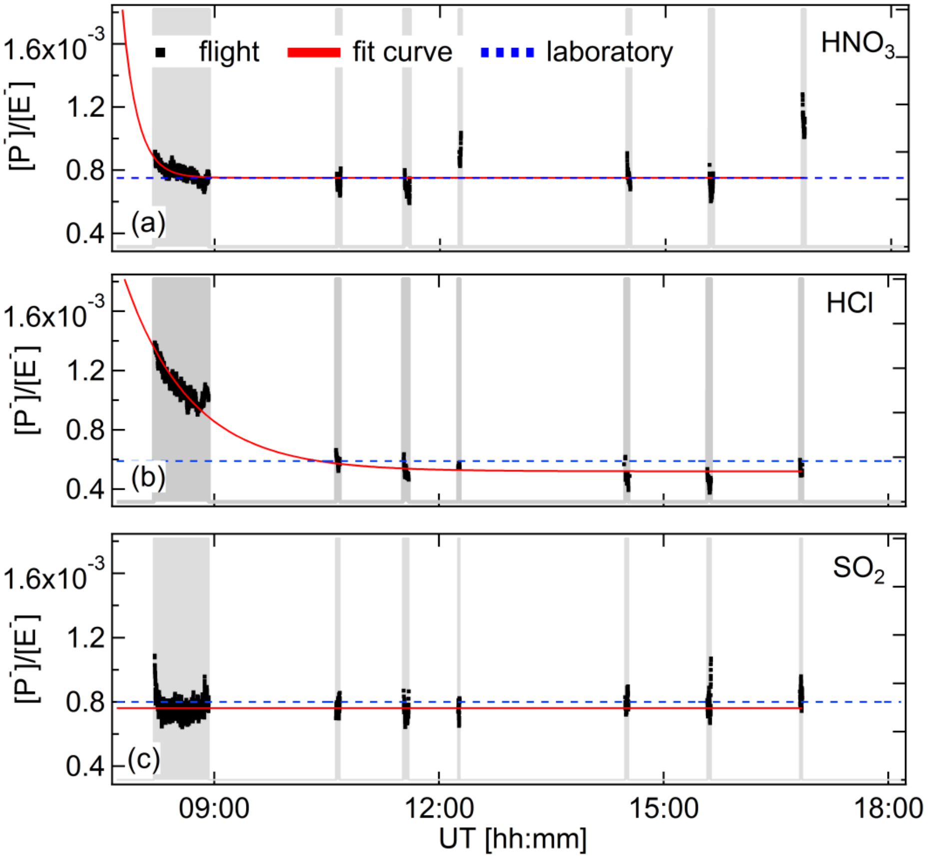

Figure 2 (Supplement Material) Ion ratios during background measurements for HNO_3, HCl and SO_2 during the flight on 11 September 2012 .Grey shaded are time sequences where synthetic air was introduced and a stable ion ratio was observed. The red curves represent the fit curves applied to correct for 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 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.

Hourly 5-min background measurements are performed by replacing the sampling flow with synthetic air. An example of such a measurement sequence obtained in tropospheric air is given in Figure 6. HCl is generally near or below the detection limit, so no decrease is visible in the background count rate. HNO_3 and SO_2 show a significant decrease. Generally after 15 to 20 s, the signal has dropped to 90% of the stable background signal anticipated by high concentrations as demonstrated in Figure 5 and tropospheric low concentrations in Figure 6. This measurement is used for investigation of background drifts. To derive an inflight detection limit, more than 5 min measurement time is generally needed in order to get a constant signal and good statistics. We have added two columns in Table 1: detection limit in flight and response time inferred from these measurements. We would like to emphasize however that the detection limits inferred from these short background measurements may be not representative due to a still variable background within the first 4 min for HNO_3.

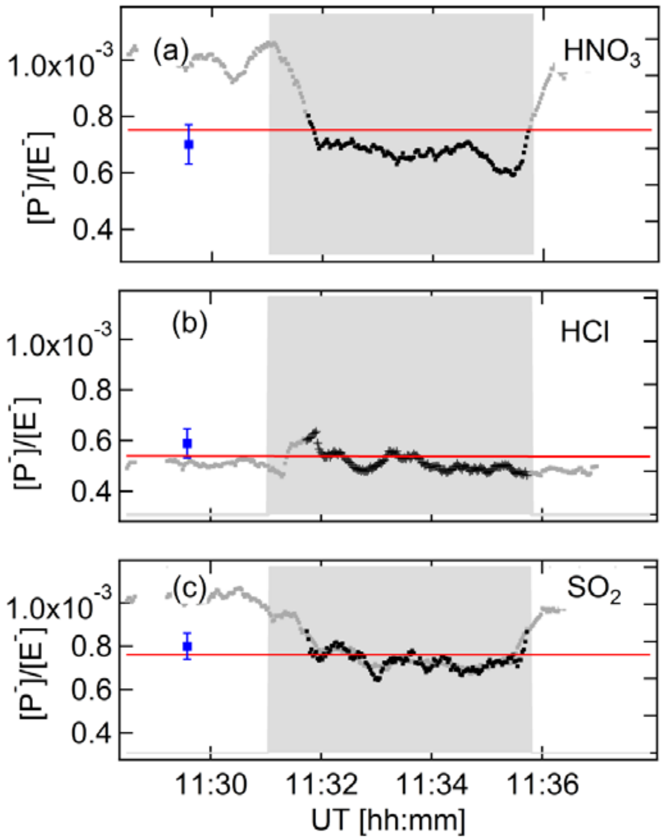

Figure 6 Ion ratios during background measurements for (a) HNO_3,(b) HCl and (c) SO_2 in tropospheric air during the flight on 11 September 2012 at 11:31 UT. Grey shaded areas show sequences where synthetic

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.

Some of the fundamental properties of the instrument are not detailed adequately. For example, what is the mass resolution of the spectrometer (give the FWHM of the mass peaks)? P13578 L 14 notes that peaks separated by 1 amu contribute less than 10% to the adjacent mass. But the results look much better than this. For example, Figure 5 shows HNO3 (m82) of a couple ppbv, and SO2 (m83) less 0.04 ppbv.
Please give a more specific and accurate description of the mass resolution. Figure 3 could help by showing only from m50-m150. Please describe the duty cycle. I don't understand how the instrument operates. How much time is spent at each mass? How much time is spent calibrating and zeroing?
We have added the following passage in section 3.1 Mass Spectrum:
*"*Generally the resolution is set to allow contamination on the neighbouring mass of less than 10%. This is achieved with a FWHM of 1 amu for mass 82 and mass 55, resulting in a low resolving power of R=m/Δm=55 and 82, respectively. The instrument has two operating modes: The mass spectrum mode scans a range from 10 to 200 amu in steps of 0.3 amu. It is recorded with a time resolution of generally 30 s per spectra every ten minutes during flight. Additionally, the instrument operates in a so-called "Hop-mode" where the ion count rate of each selected mass of interest is sequentially counted for 50 to 100 ms and then integrated for a 1 to 0.5 Hz time interval."

The paper states that backgrounds are performed hourly, but for how long? Comparisons between CIMS measurements and other measurements that integrate continuously can be complicated by the differing measurement cycles, so this detail is important. Further, it may help explain the detection limits in Table 1.
Hourly 5-min background measurements are performed by replacing the sampling flow with synthetic air. An example of such a measurement sequence obtained in tropospheric air is given in Figure 6, an example in stratospheric air with high concentrations anticipating the background measurement is shown in Figure 5.

There are an appropriate number of graphs, and they demonstrate the important points. But they could be made clearer with some simple adjustments.
Figure 1: Instead of using part numbers for the pumps, please give their pumping speed. Or, if the part numbers are important, describe them in the text. Show the important flows at the bypass and into the ion source. What is the flow into the ion source? What is the optional dilution flow used for? Either discuss in text, or remove.
The specific flow parameters have all been listed in the text. We included some in Figure 1 to give a better overview. To sum up all flows: The bypass flow is 3.6 slm, the flow through the ion source is 0.4 slm of nitrogen with additional 0.02 slm of SCF_8 *in nitrogen, the flow through the mass spectrometer is 1.5 slm. The optional dilution flow can be switched on when trace gas concentrations are so high (e.g.* SO_2 *in volcanic plumes), that the resulting product ions significantly deplete the reagent ions. To avoid non-linear effects in such a case the ambient air can be mixed with synthetic air.*

Figure 3: Plot to 1 or 10 Hz – this may give an idea of the instrument background. Why

do the 2 peaks at _M46 increase during a background? If this is unimportant, plot from 5-150.

We have split up the plot into two spectra, one tropospheric and one stratospheric, each compared to the background spectrum. Plotting the whole range seems reasonable to us since mass 19 may play an important role as a potential fragment. Additionally mass 39 (FHF^-) which is sensitive to H_2O is important to monitor potential reactions with water vapor.
Mass 46 is NO_2^-, which may be a contamination from the synthetic air bottle.

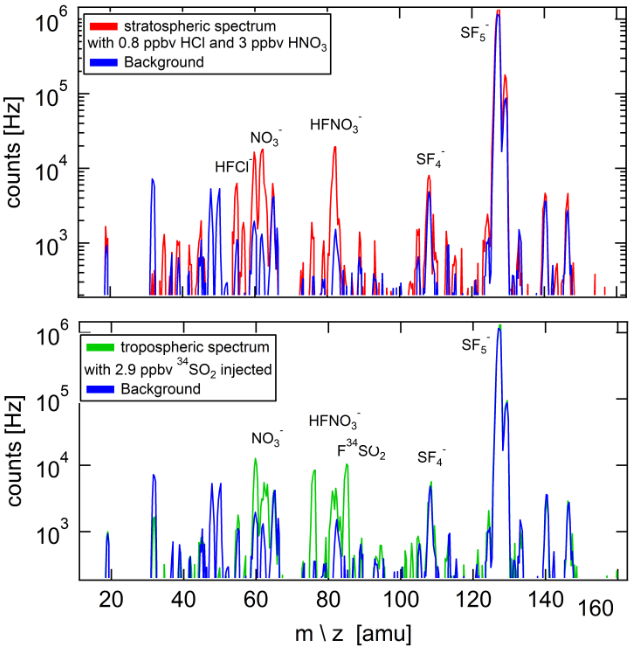

Figure 3 A spectrum of AIMS-TG obtained in stratospheric (upper panel) and tropospheric (lower panel) air south of Cape Verde on 11 September 2012 during TACTS/ESMVal is shown. The SF_5^- reagent ion at 127 and 129 amu is most prominent. Nitric acid and hydrochloric acid with their respective ions ($HFNO_3^-$ (m/z 82) and $HFCl^-$ (m/z 55 and 57)) are enhanced in the stratosphere (upper panel). The isotopically labeled SO_2 is detected as the $F^{34}SO_2^-$ ion (m/z = 85 amu) (lower panel). At m/z 146 small amounts of SF_6^- are also present. The stratospheric and tropospheric spectra are compared to a background spectrum taken while synthetic air was introduced in the inlet line.

Figure 4. In a), plot left axis from 0 to 0.1, so the linear relationship between cal and response can be seen more easily.

We have made the requested changes.

Figure 5. With log scale, plot to at least the detection limit (0.015), since there should be information there. Alternatively, use linear scale and plot to zero.

We have made the suggested changes and plotted linear scales in Figure 5.

Figure 6. Too many panels. I don't think b) is necessary. Figure 5 and 7. State time averaging of the data. HNO3>NOy at the beginning – is this a problem from measuring at higher water mixing ratios?

The data is a 20 s running mean. We have noted this in the manuscript. HNO_3 < NOy is always the case. Panel b demonstrates, that in the stratosphere the ratio HNO_3/(NOy-NO) reaches 1 which is expected and therefore an indication for the good data quality.

Smaller points:

P13568 L15: replace generating a characteristic ionization scheme with "for selective generation of reagent ions"

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."

L16 change permanently to continuously
L25 change "allow to draw" to yields
P13569 L18: change is similarly affecting to affects
L20 change in to of
L25 Huey 1998 doesn't describe aircraft measurements. Please choose appropriate reference
P13570 L5-6. I don't understand sentence. Change to "The time resolution and accuracy of CIMS can be sufficient to characterize atmospheric processes from an aircraft platform.
L22 change trace to identify
L24 Neuman 1999 doesn't describe a CIMS instrument. Please choose appropriate Reference
P13571, L18 remove while
L24 remove the
P13572 L1 change laid to place
L7 change adhesive molecules to reactive gases
L19 change continues to continuous
L15 what is the inlet temperature? L23 change "no stainless: : : is touched" to "all wetted parts are PFA"
We appreciate the many suggested improvements and comments. We have included them all in the text.

L24 what is total inlet flow? How is this measured?
As described in section 2.1 Inlet line, the bypass flow is 3.6 slm and controlled by a mass flow controller (MKS 30 slm) also described in Part 1 of the AIMS paper.

P13573 l2 remove potential, replace characterized by "accounted for"
L13 custom
L14 replace steered with drives
L21 descent
P13574 l24 Are all the ions really detected simultaneously? I think they are more likely detected sequentially. A discussion of the measurement cycle is needed.
We've added a passage to section 3.1 on the measurement modes and duty cycle.
"The instrument has two operating modes: The mass spectrum mode scans a range from 10 to 200 amu in steps of 0.3 amu. Mass spectra are recorded with a time resolution of generally 30 s every ten minutes during flight. Additionally, the instrument operates in a so-called "Hop-mode" where the ion count rate of each selected mass of interest is sequentially counted for 50 to 100 ms and then integrated for a 1 to 0.5 Hz time interval"

P13575 l4 The reduction of the HNO3 background is an important advancement. The paper should quantify this reduction and compare with published values of instrument backgrounds in order to highlight this improvement.
Unfortunately a polonium ion source, to which it should be compared, is not available, so a direct comparison of the two backgrounds with one instrument is not possible. From the experience with the polonium ion source where HNO_3 *was produced in the ion source we agree that the discharge source is definitely a significant improvement, given that the background can be reduced by lowering the applied voltage. Additionally the addition of* NH_3, $NaHCO_3$ *or other reactive components is not necessary.*

l11 replace instantly with rapidly
P13576 l5. I don't understand sentence. Do you mean that sensitivity to HONO is the same as HNO3? Or within an order of magnitude?
We used the same sensitivity for HONO as for HNO_3*.*

P13577 l3 remove "reducing the reaction efficiency, e.g."
P13579 l15 I don't what reservoir tank means.
We have replaced the word with "cylinders"

L21 remove in a defined manner
We have made the suggested changes.

L23 is nitrogen mass flow controlled?
L24 is pressure in permeation ovens measured? How much does it vary with altitude?
It is mass flow controlled by a critical orifice made of glass which is located downstream of the oven. The N_2 *pressure in the oven is set to a constant pressure of 2 bar with a pressure regulator. This allows a defined* N_2 *flow through the orifice of 90 to 100 sccm.*

L28 change to "the pressure inside the oven is independent of the inlet pressure"
P13580 l3 You need to know the total inlet flow to determine calibration mixing ratios. What is it?
C4835 L9 what is at 3 bar? Earlier, perm ovens were said to be held at 2 bar. Flow through perm oven isn't important – instead, give total inlet flow.

The pressure in the permeation oven is set to 2 bar above 1 atmosphere, thus in total 3 bar. We have included the flows in Figure 1 and in section 2.1 "Inlet line". We agree that the flow through the permeation oven isn't important since it only comprises a small dilution of the inlet sampling flow (~0.5%) but the pressure in the oven should be kept constant for this may affect the permeation rate (See Arnold et al., 2007).The total flow is 4.7slm.

L10 remove standardized to. And is this the total HCl concentration, or the concentration for the 35Cl isotope?
This is the total concentration but we only compare it to mass 55 ($HF^{35}Cl^-$*) for the calibration, assuming a constant isotope ratio in the permeation device as in the atmosphere.*

L28 Description of calibration is confusing. The point of calibrating is to account for inlet and other losses, and the important parameter is the calibration mixing ratio in the inlet. How does flow through dilution oven and inlet loss affect the inlet calibration mixing ratio?
The largest uncertainty of the calibration comes from the permeation rate. However the mass flow controller controlling the bypass flow and the flow through the mass spectrometer adding up to the total sampling flow additionally contribute to the uncertainty of the calibration. For the mass flow controller we assume an uncertainty of 8 % and for the gas flow through the mass spectrometer of 5 %.

13581 l1 to be 12-%
l2 remove thermal. The ovens should reach thermal equilibrium very rapidly. Changing concentrations are probably from surface effects.
L3 replace it is pursued to remove it with they are removed
L4 is flow also kept constant?
L19 replace inevitable rest amount with impurity
L20 delete any kind of

P13582 l29 replace strong with large
We have made the suggested changes.

13583 l9 How do you know the reaction rates for the differing isotopes? Please reference
Due to the higher mass of deuterium the zero-point energy is lower. Therefore the activation energy for reactions with deuterium molecules is decreased compared to equivalent bonds with hydrogen (P.W. Atkinson and J. de Paula. Physikalische Chemie. Wiley-VCH, 2006.). The so called kinetic isotope effect is largest for deuterium and hydrogen, due to the large mass difference but it is negligible for the sulfur isotopes.

P13584 section 5: the mass spectrometry tutorial isn't necessary in this instrument paper. Instead, reference earlier work (e.g. Arnold or Viggiano from early 1990s), and this section can be shortened or entirely removed
We have made the suggested changes and shortened the first part of the section.

P13586 l17. Why can't noise be determined just as well in flight? This seems particularly important, since part of the value of this paper is demonstrating operation on a high altitude aircraft at reduced pressure and reduced water
We try to assess this comment with a separate column in Table 1 and an additional plot (Figure 6) comparing the background during flight with the laboratory background. Background levels generally agree. Noise determination in flight means losing valuable measurement time. Short background checks of 5 min may be useful to determine the background drift but not the detection limits since most of the 5-min time interval is used to flush the inlet walls. Therefore the detection limits inferred in flight are only meaningful for large concentration drops down to the detection limit. We have also remarked this in the text.

P13587 l4 For what count rates is this true? The statistical noise should go as square root of number of counts. I am not clear what noise the discharge ion source adds. Is it a constant count rate at all masses?
We assessed the change in the ion ratio and noise with increasing ion source voltage in the supplement material Figure 1. The plot shows the behavior for $HFCl^-/SF_5^-$ *which is the general behavior for most masses. With increasing ion source voltage the background signal as well as the standard deviation which is directly linked to the detection limit increases by a factor of more than 3 between 400 and 3500V applied to the needle tip.*

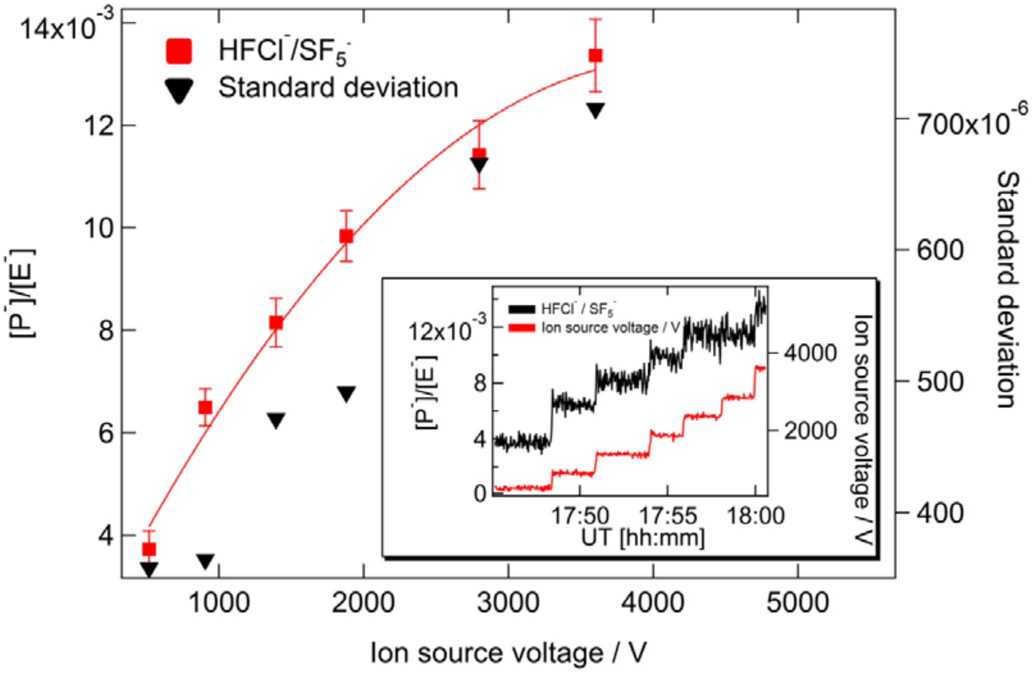

Figure 1 The ratio of product to reagent ions [P$^-$]/[E$^-$] (here HFCl$^-$/SF$_5^-$) during background measurements is shown for different ion source voltages. The inset shows the timeline of the signal, while the larger plot shows the direct correlation of the average values of [P-]/[E-] and the standard 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.

L23 please explain how and why you use the nylon filter backgrounds. Do they agree with dry air flushes?
The nylon filter was originally intended for background measurements in humid mid tropospheric conditions. Since we flew only in upper tropospheric lower stratospheric dry air, the nylon filter was only used once in order to test it. Additionally, as mentioned by the reviewer, some substances are scrubbed less effectively than others; therefore the application is optional and limited to very specific flight conditions.

P13588 l2 from the calibration procedure
L15 replace despite with although
We have made the suggested changes.

L20 this is an interesting idea to passivate the inlet lines with HNO3. How long does this passivation last? Has this been tested (I wasn't sure from the text)? This must also increase the backgrounds. Further, if inlet passivation changes HNO3 transmission, the instrument may have a nonlinear response, where HNO3 transmission increases when HNO3 mixing ratios are larger. A figure that shows improved time response before and after passivation would be helpful, or at least this statement should be supported by some quantification.
We changed the text to
"Passivation is achieved by flushing the calibration line on the ground before take-off with the HNO_3 *and*

HCl and may improve the reaction time of the calibration."
Passivation of the inlet can only qualitatively be discussed and is prone to many other uncertainties like memory effects. The calibration lines are less affected by atmospheric conditions during flight (e.g.humidity) but they contribute to the time response of the calibration. The original idea was to passivate the inlet with isotopically labeled $H^{15}NO3$ *but has never been put to practice in flight due to the interference with* SO_2 *on mass 83.*

P13589 l7 How do you account for the changing sensitivity with water? Line 26 states that water vapor dependencies can be corrected. Are all the data corrected for water?
We mainly correct the water vapor dependent background signal which is monitored after the flight on the ground with water vapor concentrations from 50-250 ppmv. Sensitivities during TACTS/ESMVal did not show a significant dependence on water vapor since only dry UTLS conditions were sampled. We therefore refrain from showing a water vapor dependence of the measurements.

P13590 l1 I'm not sure what is meant by occasional evaluation of measurements at higher water vapor concentrations. Please be clearer about the range of water vapor that can be sampled. Perhaps discuss the utility of a dilution flow as shown in Figure 1 (if the dilution flow can be used to sample at high water vapor).
The dilution flow ranges between 0.5 and 5 slm. Thus between 5 and 90% of the total flow can be diluted without adding additional uncertainty to the measurement. Measurements at lower levels with high water vapor concentrations and higher trace gas concentrations in the degassing volcano Etna are described in detail in Voigt et al., 2014. In this paper, the sensitivity was tested for higher humidities. During TACTS/ESMVal we mainly focused on the UTLS measurements and omitted data for $H_2O > 100$ *ppmv.*

L23 what is the water vapor concentration at 4 km altitude?
H_2O *mixing ratios at 4km can be very variable between a few 100 and more than 1000 ppmv. In this case* H_2O *at 4 km was 200ppmv. AIMS data were discarded below 8 km.*

P13591 l3. Were measurements really obtained in a thunderstorm cloud? And why doesn't HNO3 change in the cloud?
The measurements were obtained in clear air after a thunderstorm has already passed. Primary products of lightning, similar to combustion, are NO and HONO. HNO_3 *is a secondary product and thus not built up in the same way and time scales. It may also have been removed through wash out effects. Therefore a significant increase of* HNO_3 *in the freshly injected, NO-rich air is not expected. Shortly after reaching the flight level nitric acid is enhanced. This may represent converted NOx from an aged outflow.*

L13 what is NO2 (calculated)?
During daylight, NO_2 *is mostly photolyzed, thus near zero.*

L17 how is the time from the lightning event known? Does this factor of 7 difference C4837 assume only photolysis and no dilution at all?
Dilution calculations are not required for the ratio of HONO to NO since both substances dilute equally. Therefore only photolysis is assumed.

L29 I don't know what is meant by the derivatives of the stratospheric tracers. Fewer panels here may be less confusing.
We've changed the wording to stratospheric tracer ratios.

P13592 – nearly all of this discussion can be removed. The use of correlations of

tracers to understand atmospheric sources is well established. The discussion of NOx and HNO3 as markers of air mass age has appeared in many papers (e.g. Bertram et al., Science 2007). I recommend removing this material to shorten the manuscript.
We would prefer to keep the plot and add a few sentences on how data quality can be demonstrated also from the atmospheric measurements and the tracer-tracer correlations. In any case, the analysis shows potential applications of the data in a scientific context, which we think is a necessary requirement of a new instrument. It is also a good demonstration of the performance of the instrument.

"The tight correlation of HCl and HNO_3 suggests a major influence of stratospheric air, however with significant contributions from tropospheric tracers like NO. The high correlation coefficient of R^2=0.979 in stratospheric air additionally demonstrates similar response times of the instrument with respect to both substances. Memory effects are not affecting the compact correlation of HNO_3 with the less reactive gas HCl. The ratio of NO/NOy decreases with distance to the tropopause. The ratio of NO/NOy also acts as a chemical clock. Nitrogen oxides are injected in the atmosphere as NO that is subsequently converted to other NOy species e.g. HNO_3. Therefore a low NO/NOy ratio indicates an aged air mass that is not subject to recent NOx emissions. In the unperturbed stratosphere, NOy is dominated by HNO_3, indicated by the blue data points. The values of 1 for the HNO_3/(NOy-NO) ratio in the stratosphere are in agreement with Neumann et al., 2001, demonstrating the reliability of the data. While HNO_3 has tropospheric and stratospheric sources, the ratio of HNO_3 to NOy indicates the age of the air mass, similarly to NO/NOy.

P13593 l12 custom
L17 replace allow to identify the origin of: : : with "enable identification of air mass origin"
P13594 l8 remove naturally and e.g.
References: several typos
We have made the suggested changes.