Our response/reactions are in black and comments of the reviewer in blue

General Comments

The paper reports results from the ATTREX campaign performed in the Eastern Pacific during 2013, which probed the UT, TTL and LMS. The experimental results are of high importance for determining the overall fraction of inorganic bromine injected to the stratosphere, i.e. capable of destroying ozone in the LMS. BrO measurements are compared with modelled output from the TOMCAT/SLIMCAT model, which runs at high resolution with the specific meteorology (i.e. ECMWF Era-Interim reanalysis) present during the ATTREX campaign.

Even when the presented results possess a notable scientific impact as they probe inorganic bromine species in a region of major importance for constraining stratospheric bromine injection, the presentation of results is in this reviewer´s opinion rather disordered, and the description of the model configuration/output used to interpret their measurements is rather confusing. Therefore, several issues, both structural/technical and scientific/descriptive, must be dealt with before this MS is ready for publication. I recommend the following major comments be addressed before this paper can be accepted in ACP.

We are very grateful for the time the reviewer spent on reviewing the manuscript. The comments will help us to improve the manuscript. Please find below our point-to-point responses to the comments and queries.

Major Comments:

1. The abstract is too long and contains unnecessary information such as i) the instruments used for probing the composition of the tropical atmosphere, ii) a description of the minor deficiencies in the modelled transport, iii) a reference to a paper with the definition of the TTL. Also, the most important information in the abstract is quite disordered and the main results (i.e., measured numbers which are the major findings of this work) are only given in the very last lines.

We agree that the abstract was too long and shortened it by 8 lines. However, we disagree that it contains unnecessary information since the description of i) is relevant for the interpretation of our measurements, ii.) is honest in that the modelled curtains need a (joint) vertical adjustment in order to match the observations, iii.) since conflicting definitions of the TTL (in particular its bottom) appear in the literature.

2. The authors used a quite simple proxy to separate tropospheric air from stratospheric air: the condition of [CH4] being larger or smaller than 1790 ppb. This selection is neither justified nor referenced within the MS, and its use should be clearly justified as many of the forthcoming results depend on the validity of this assumption. Indeed, the histograms shown in Fig. 13 seem to contradict the validity of the CH4 proxy in splitting tropospheric from stratospheric air for theta > 390 K. Also, within the abstract and text, the CH4 condition is defined both respect to 1390 and 1790 ppb, introducing an additional inconsistency to the definition.

The [CH4] = 1390 ppb criteria were typos. They should have read [CH4] = 1790 ppb. We corrected these mistakes in the text. Our intent was not to separate tropospheric from stratospheric air since only air from subtropical LMS and TTL was sampled. The [CH4] >< 1790 ppb criterion is used to (roughly) separate younger air sampled within the TTL from relatively older air from the subtropical LMS. No other more robust method was available from the aircraft observations.

3. In the abstract and methodology, the authors declare that the TOMCAT/SLIMCAT model was "constrained to the measured O3 and NO2 and adjusted to match the observed concentrations of some brominated source gases" (P2,L6-7). But no detail is given in the methodology about how this

special configuration was applied in the model. Later, many of the results are interpreted and discussed based on the output obtained from the global model. In light of the importance of the observations and conclusions drawn here, mostly the inferred Bryinorg, the authors should describe specifically how the model was constrained or adjusted.

The method how to match the observed O3 (and CH4) is described I detail in Stutz et al., (2016). In short, the following: TOMCAT/ SLIMCAT has a height resolution of about 1 km in the TTL. The GH on the other hand, mostly flies at altitudes located between the model grid points. Therefore, an interpolation is necessary to account for sub-grid trace gas concentrations and to allow for an accurate comparison between the model results and the measurements. Moreover, in order to compensate for the models small (and varying) inaccuracies in describing the vertical air motion in the atmosphere (c.f., due to transient gravity waves), the modelled concentration curtains of all chemical tracers are jointly shifted in the vertical (by typically less than modelled vertical grid) until measured and modelled O3 agree. Once this vertical adjustment is performed (e.g., see Stutz et al., 2016, compare Figure 12 with 13), the measured and modelled CH4 (a transport tracer measured by UCATS and HUPCRS) and NO2 (a photochemical active species measured by the DOAS instrument) largely agree (see Figures 4 to 9). As a consequence, it can be concluded that (a) the vertical adjustment is reasonable and justified (from the agreement seen in CH4), (b) that the scaling method works well for NO2 and hence for BrO, and (c) that the adopted NOx/NOy photochemistry is able to reproduce measured NO2 (see Stutz et al., 2016).

We changed the following parts of the manuscript in order to clarify :

P1 line 16 cont: Depending on the photochemical regime, the TOMCAT/SLIMCAT simulations tend to slightly under-predict measured BrO for large BrO concentrations, i.e. in the upper TTL and LS.

P11, line 16: The text now reads…… The panels (b), and (c) of Figs. 3 to 8 show comparisons of measured and modeled CH4, and O3 mixing ratios. Here the measured and modeled species reasonably agree within the given error bars, after the modeled curtains are altitude-adjusted by the same amount until measured and modelled O3 agree (for details see Stutz et al. (2016)). Noteworthy is, that in most cases the altitude adjustment is less than the grid spacing of TOMCAT/SLIMCAT (about 1 km in the TTL), thus mostly accounting for the altitude mismatches of the actual cruise altitude of the Global Hawk and the model output rather than deficits of the model to properly predict the vertical transport.

For example:
My major concern is about how removal/washout is considered in the model, mostly within the UT and TTL, as these processes will control the overall Bryinorg burden in that region of the atmosphere. The only reference to removal rates in the MS that I could found was on Page 15: L26 "…, and assuming no bromine is effectively lost in the troposphere, …" and L30-31 "Therefore effective loss processes for inorganic bromine, for example by heterogeneous uptake of inorganic bromine on aerosol and cloud particles, must act in the atmosphere". Detailed information on the removal processes considered here for brominated species should be given and also how they affect the BrO/Br ratios.

First, since the measurements were solely performed in the LMS and TTL, details in the budget and photochemistry of bromine in the troposphere only need to be considered in the model in as much as they are relevant to the budget of bromine in the TTL and LS. A description how the model is constrained to the budget of bromine in the troposphere and what processes are considered is given in section 2.7. In order to account for more complicated bromine sources (e.g., derived from sea salt and/or yet unknown minor organic brominated species), an additional 0.5 ppt of BrO is added to the near surface bromine, but only where TOMCAT/SLIMCAT predicts [BrO] < 0.5 ppt). Further, this modified BrO curtain is only used for the RT simulations, in order to better represent the BrO

absorption introduced by the small fraction of light traveling through the lower troposphere before being analysed by the limb oriented telescope on the GH. This approach is justified based on (a) the findings reported in Stutz et al., Figure 8b, and 15, (b) the findings of Volkamer et al., 2015 on BrO in the tropical tropics, (c) Dorf et al., 2008, and our earlier work (Harder et al., (1998), Fitzenberger et al., (2000), (d) Chen et al. (2016), and (e) our measurements around Borneo during SHIVA. This assumption also appears to be justified based on recent modelling studies which argues that sea salt derived bromine might be delivered to the TTL (e.g., Schmidt et al., JGR, 2016 see also below).

However, we re-call here again that in the measurement/model inter-comparison (sections 4.1, 4.2 and 4.4) measured/inferred quantities are directly compared with the TOMCAT/SLIMCAT model output, except!!! that all modelled curtains are vertically shifted by the same amount until measured and modelled O_3 agree. The latter is necessary since (a) to avoid interpolation errors between the actual measurements altitude, the measurements themselves, and the model predictions provided in discrete step which do necessarily correspond to the altitude of the measurements, and (b) to compensate for small mismatches in the actual and modelled vertical transport. Mostly (> 98%), the necessary vertical shifted than the vertical spacing of the model (~1 km).

Accordingly, and to account for the omitted sea-salt source (and in order to avoid any confusion) we (1) changed our text (page 8 line 25 cont.) to …

No other (cf., unknown organic or inorganic) sources of bromine for UT, LS, and TTL are assumed (e.g., Fitzenberger et al. (2000), Salawitch et al. (2010), Wang et al. (2015)). Omitting the release and heterogeneous processing of bromine from sea-salt aerosols (e.g., Saiz-Lopez et al. (2004)) in the model for the sake of saving computing time appears justified since (1) even though it is predicted to be relevant for bromine (~30% of the total Br_y^{inorg}) in the free troposphere (Schmidt et al. (2016)), its contribution to BrO in the TTL is at most of the order of the accuracy (~0.5 ppt) of our BrO measurements, (2) its time and space dependent sources (as for the brominated VSLS) are not well constrained, (3) in the modelled troposphere inorganic bromine only serve as boundary condition for bromine in the TTL, and (4) the additional BrO would not affect the BrO measurements-based calculation of Br_y^{inorg} for the TTL (see below).

(2) added the following in the discussion (section 4.4, page 14, line 35 cont.)

This gap could partly be closed by adjusting the CH2Br2 surface concentration and atmospheric lifetime, or by considering a detailed scheme for dehalogenation of sea salt, i.e. bromine activation (e.g. Saiz-Lopez et al. (2004), Fernandez et al. (2014), Schmidt et al. (2016)). Adjusting CH2Br2 would add 0.4 ppt of Br_y^{inorg} , or ~ 0.3 ppt to BrO, thus removing the flight-to-flight scatter in source gas concentrations (~ 0.8 ppt) in Br_y^{inorg} . This could for example be done by a detailed back trajectory and source appointment analysis to which a forthcoming study will be devoted. Likewise, release of sea salt halogens to the gas-phase could add another 0.5 ppt to BrO (or about 0.7 ppt of Br_y^{inorg}) in the upper TTL (Schmidt et al., 2016).

and
(3) changed text in the introduction (page 2, line 11 cont).

…(3) so-called very short-lived species (VSLS), and (4) inorganic bromine transported into the upper troposphere, e.g. previously released from brominated VSLS and/or sea salt (e.g., Saiz-Lopez et al. (2004), Fernandez et al. (2014), Schmidt et al. (2016)). This inorganic bromine is also transported into the stratosphere.

(4) On page 14, line 23 cont.: Therefore, effective loss processes for inorganic bromine, for example

by heterogeneous uptake of inorganic bromine on aerosol and cloud particles, must act in the atmosphere (e.g., Schmidt et al. (2016)).

b. Also, In the model description (P8,L21-23), the surface concentration of VSL is 1.00 pptv for CHBr3, CH2Br2 and other VSLS. Why at (P12,L12) a value of 1.05 pptv is informed?. Further on, in the conclusions (P17,L15) the 1.0 ppt value is mentioned again. It may simply be a typo? Even when a 0.05 ppt value will not make a difference, this point should be made clear and consistent. How were the surface emissions adjusted?

Thanks for finding this error, we corrected the typos on P12 and P17.

4. In relation with my previous comment about an improved description of the specific model configuration used in this study, the authors state that "No other (c.f., unknown organic or inorganic) sources of bromine for UT, LS, and TTL are assumed (e.g., Fitzenberger et al. (2000), Salawitch et al. (2010), Wang et al. (2015), and others), except that we add 0.5 ppt to the modeled tropospheric BrO in agreement with the finding discussed below (section 4.6)". Later in Section 4.6, no specific mention is given about this additional source of BrO.

The reviewer is correct this was not clearly stated. As explained already above, only in the simulated curtains used for the RT calculations BrO was set to 0.5 ppt in the lower troposphere when the modelled predicted lower BrO. This was done in order to better represent the BrO absorption introduced by the small fraction of light traveling through the lower troposphere before being analysed by the limb oriented telescope on the GH (at 14 to 18 km) (see Figures 14 and 15 in Stutz et al., 2016).

We accordingly changed the text in section 2.6 (page 8, line10 cont.) to: "Only in the RT simulations [BrO] is set to 0.5 ppt near the ground, where TOMCAT/SLIMCAT predicted lower BrO concentrations (see Figure 2 middle right panel), in agreement with the findings discussed in Stutz et al. (2016) and the recent study of Schmidt et al. (2016). "

As BrO is used to constrain inorganic bromine using TOMCAT/SLIMCAT, a clear description of this additional source of BrO must be given in the text.

No, TOMCAT/SLIMCAT is freely run with the sources of bromine as given in section 2.7.

b. Yang et al., 2005 and Ordoñez et al. 2012 show that an additional source of inorganic bromine from sea-salt in the MBL is required to reproduce observations. Fernandez et al 2014 highlighted the importance of the sea-salt contribution for Bry in regions of strong convection such as the tropical western-pacific. Is this additional source of BrO in this work related to sea-salt recycling?, if so, is it constrained as a boundary condition or explicitly calculated?

Please see our response to previous questions

Please expand the discussion about this important omission. In Section 4.6 (P16,L10-13) the impact of sea-salt (or any other additional source) on Inferred Brytotal should also be discussed.

The recent modelling study by Schmidt et al., JGR (2016) indicates that 30% of inorganic bromine in the free troposphere might be due sea to salt. Their model also simulates [BrO] concentration of about 0.5 ppt in the free tropical troposphere. We find [BrO] < 0.5 ppt from the down-looking observations (Figure 15 in Stutz et al., 2016), and that between 0.5 ppt to 5.25 ppt (mean 2.63 +/- 1.04 ppt) of inorganic bromine is transported through the bottom of the TTL. Consequently, our conclusion on contribution 4 (which includes sea-salt derived bromine) in the tropical troposphere appears to be fairly reasonable. Since SLIMCAT/TOMCAT is a more stratospheric centric CTM, omitting details of heterogeneous and microphysical processes of sea-salt derived bromine in troposphere, i.e. treating it as a model boundary condition, is justifiable to save computer time. Finally, since in our assessment

on TTL bromine is based on measured BrO (CH4, O3, and NO2) and detailed photochemical modelling, no sizeable amount of bromine is omitted in the model, except that eventually some bromine (sub ppt, see below) might be tight to the TTL aerosol (which however is not really founded based on the findings of Murphy et al., 2016, (Halogen ions and NO+ in the mass spectra of aerosols in the upper troposphere and lower stratosphere, Geophys. Res. Lett., 27, 3217–3220, 2000 and our personal communication in 2000).

For the changes to the text see above.

5. In Section 2.1 (DOAS measurements of O3, NO2 and BrO), a companion paper (Stutz et al., 2016), describing the DOAS measuring technic used during the ATTREX campaign, is introduced. However, the current manuscript makes too many references to Tables, Figures and Sections within the Stutz et al. paper, which do not introduce additional clarification and in most cases difficult a direct reading of the main results. Please, revise the whole manuscript on this respect and keep the references to the Stutz et al., 2015 only when they are relevant for the results presented here.

In fact, we were trying to fit the entire material into a single manuscript, i.e. (a) a description of the instrument, (b) the spectral retrieval (DOAS), (c) the novel technique (scaling method) for the concentration retrieval, (d) the RT and CTM calculations and (e) the data interpretation. However combining (a) to (e) would have made a single manuscript too long and thus rather challenging to read. Consequently, we decided to split the study into two papers. We recommend to read the Stutz et al. (2016) paper in order to understand what information can be inferred from the measurements, and what information can not be obtained (c.f., averaged profiles of measured radicals, or a direct comparison to the Fernandez et al., 2014 and Saiz-Lopez and Fernandez, 2016 studies because they address a different atmospheric compartment or altitude range, see your comment below). Because the present manuscript often refers to the measurements, the underlying retrieval, and data interpretation, we feel it is rather necessary to frequently refer to the Stutz et al., (2016) manuscript. We would also like to add that the two manuscripts are linked to each other in the AMT/ACP environment and should be considered part 1 and 2 of the same study.

6. The results presented here somehow contrast with previously published measurements/modelling approaches. While the discussion respect to the findings from Wang et al., 2015 and Volkamer et al., 2015 are extensively discussed in section 4.3 (P13,L3-26), just a brief comment on the discrepancies respect to Fernandez et al., 2014 and Saiz-Lopez and Fernandez, 2016 is presented (Section 4.5, P15,L0-4).

The definition of the lower end of the TTL remains controversial, as already discussed above. We had several discussions with colleagues on this topic but no consensus could be reached. In fact, there is only little overlap (with respect to altitude, Θ, p) between the BrO measurements of Wang et al., (2015) Volkamer et al., (2015) and modelling presented by Fernandez et al., (2014) and Saiz-Lopez and Fernandez (2016) (which according to our preferred TTL definition based on Fueglistaler et al. (2009) addressed more the bromine photochemistry in the upper tropical troposphere) and our study. Since two different regimes are described it is not necessary to address them in greater detail (except to compare Wang et al., (2015) and Volkamer et al., (2015) with our BrO profile (see Figure 3a).

With regards to the [Br]/[BrO] ratio, the common pattern shown in Figs. 4-9 is that whenever BrO mixing ratios decrease, both HBr and atomic Br increase.
Yes, this is a consequence of the adopted JPL bromine photochemistry in TTL and LS and decreasing O3 concentrations towards the (our) bottom of the TTL.

When this inversion is observed, the flight altitude and O3 levels also decrease, indicating that the lower TTL is being probed. In most cases HBr surpasses Br, while at some points (e.g., Fig 5f, 23:00) [Br] dominates. Thus, the Br and HBr prevalence seems to depend on the height (and possibly temperature,

not shown) at which the TTL is being probed. I would suggest further discussion and interpretation of those results. Particularly, if the absence of heterogeneous recycling is affecting the inferred BrO/Br ratio?.

We added two small paragraphs to clarify this issue.

P12, line 5 cont: Panels (e) in Figures 3 to 8 compare measured and modelled BrO. Again measured and modelled BrO mixing ratios reasonably compare for most of the flight, but sizable discrepancies are also discernible for some flight sections. Possible reasons for the latter, which are discussed in the following section, may be due to deficits in the models assumption of the sources of bromine (see section 4.2), and/or deficits in the adopted photochemistry (see section 4.4).

P14, line 5 cont: Overall this behavior is expected from arguments based on the amount and composition of the brominated organic and inorganic source gases, their lifetimes, atmospheric transport, and photochemistry (e.g., Fueglistaler et al. (2009), Aschmann et al. (2009), Hossaini et al. (2012b), Ashfold et al. (2012), WMO (2014), Fernandez et al. (2014), and Saiz-Lopez and Fernandez (2016), and others). In particular, for our measurements at daytime, it is observed that (a) BrO increases with O3 and available Bry^{inorg} and thus altitude, (b) the predicted BrO/Bry^{inorg} ratio decreases towards the bottom of the TTL, where (c) HBr and/or Br atoms may become as abundant as BrO, but HOBr does not play a major role in the Bry^{inorg} partitioning. While observation (a) is primarily due to the increased destruction of the short-lived Bry^{org} species and the efficient reaction of the released Br atoms with altitude dependent ozone concentrations, observations (b) and (c) are due to reactions of the Br atoms with CH2O, (and to a lesser extent H2O2) into HBr which is recycled to Br atoms through reactions with OH and on the available surface of aerosols and cloud particles, as predicted by Fernandez et al. (2014), and Saiz-Lopez and Fernandez (2016). Noteworthy is also the predicted minor role of HOBr formed by reactions of OH radicals with heterogeneously produced Br_2, or by the reaction HO2 + BrO. While the rate of the former reaction is small due to short photolytic life-time of Br2, the rate of latter reaction is small due to the small HOx concentrations in the TTL compared to photolysis of HOBr at daytime.

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7. In section 4.1 the authors directly start showing results for Figs. 4-9. Not a single description is given to Fig. 3. Thus, Fig. 3 should be moved further down in the text.
We agree. Figure 3 is now moved to become Figure 12.

In P11,L17-20 you state that “The excellent agreement achieved between measured and modelled CH4, and O3 lends confidence that the altitude-adjusted TOMCAT/SLIMCAT model fields reproduce well the essential dynamical and photochemical processes of the probed air masses.”. The authors can only assure a sentence like this by means of a full vertical profile validation of the species, not only comparing data at a specific level.

In the core we agree, but as matter of fact our observations are covering largely different altitudes (for example during the dives), photochemical regimes (the LMS, and TTL), SZA’s ranging from 0^o to 80^o degrees et cetera that at least we were really surprised how well the measurement and the model agree. Accordingly, we replaced excellent with …..astonishingly good …. in the text.

8. The works possess several informal phrases which may not fit well within a scientific work. I list below some of them (but certainly not all), that maybe should be rephrased to a more appropriate structure, justified by numbers/references or removed from the MS.

Abstract, P2,L4: “… and the expectation based on the destruction of brominated gases.”

We accordingly changed the phrase to …. and the expectation based on the photochemical destruction of the brominated source gases.

P4,L1: "… indicating that some Bry_inorg (i.e. several ppt) is directly transported from the tropopause"

We accordingly changed the phrase to … "… indicating that variable amounts of Bryinorg (i.e. several ppt) from the troposphere into the stratosphere"

P9,L17: "…, some exciting observations and details"

We accordingly changed the phrase to "…, some observations, their details as well as some results"

P11,L4: "… we refrain from this much more complicated approach"

We keep it since as conclusion (using a personal pronoun) the phrase appears to be justified.

P11,L27: "… NO2 concentrations meet the expectations for NOx"

We accordingly changed the phrase to …."… NO2 concentrations meet the expectations with respect to it's a partitioning and total NOx"

P16,L24: "By far …"

We accordingly erased …By far …"

9. There are several other sentences that may benefit from revision. I list some of them below:

P8,L22: [CHClBr2,CHCl2Br,CH2ClBr,…]. What the three dots means? Please specify.

In English grammar it is called an astrophic continuation, see https://en.wikipedia.org/wiki/Glossary_of_literary_terms i.e. it stands for other not considered minor brominated organic source gases. In fact the native speakers in list of authors had no objection to use it, but it could be necessary to contact an Anglist on this.

P12,L3: NOy=(NOx,N2O5,HNO3,HO2NO2,…. What the three dots means? Please specify.

See above. Other, not considered minor NOy species for example ClONOy and BrONOy.

P17,L14: Once again [CHClBr2,CHCl2Br,CH2ClBr,…]

See above. Other, other not considered minor brominated organic source gases.

P18,L3: (Here: cite Hossaini 2016, acp when published)?

Thanks. We updated the references to Hossaini et al. (2016).

Figures Comments:

Fig. 1: All results shown in this paper are for the ATTREX Flights performed during 2013 in the Eastern Pacific. Then why do you show panel A with the ATTREX 2014 flights that are not used here?

We accordingly changed the Figure to only display the 2013 flights.

Fig. 3: This Figure should be moved down in the text to make it consistent with the presentation of results. Also, it may be unified into a 2 panel figure, with only 1 caption that distinguishes between the a) and b) panels. The same applies for Fig. 13.

We moved both Figures to become the new Figures 12a and b, and we merged them into a two panel plot. We also merged Figure 13a and b.

Fig. 4: A portion of the caption is completely missing.

Figure 3. Panel (a) shows the time-altitude trajectory of the sunlit part of the GH flight track (SF1-2013) on Feb. 4/5, 2013 (SF1-2013). Panels (b)-(e) show inter-comparisons of TOMCAT/SLIMCAT-simulated fields with observations of (b) CH4 (UCATS), (c) O3 (NOAA), (d) NO2 (mini-DOAS), and (e) BrO (mini-DOAS). The grey-shaded error bars of the mini-DOAS NO2 and BrO measurements includes all dominating errors, i.e. the spectral retrieval error, the overhead and the error due to a tropospheric contribution to the slant absorption, and the absorption cross section uncertainty. Panel (f) shows the SLIMCAT modeled Bry partitioning for the standard run #583. Panel (g) shows a comparison of inferred and modeled Br_y^{inorg}, including the errors as a grey band. The dashed vertical lines in Figures 4-9 separate different atmospheric regimes: (I) is the extra-tropical lowermost stratosphere, (IIa, IIb, ..) different mixing regimes of air from the extra-tropical lowermost stratosphere, and (III) from the tropical tropopause layer.

This should be carefully controlled before submission. Also, following the equivalent figures for the rest of the flights (Figs. 5-9), no mention is given to what the vertical dashed lines represent. In addition, it would be very helpful to show a constant (dashed) line for O3 = 150 ppb and CH4 = 1790 pptv to distinguish the periods where the subtropical and TTL air was being probed.

Wouldn't it be a good idea to show the "mean temporal profile" measured by all of the ATTREX flights (i.e., for equivalent SZA)??

We thought about it and came to the conclusion that this is not really a good idea because the amount of BrO would also depend on total Br_y^{inorg}, O3, NO2, ….

Fig. 13: The large values of BrO at theta = 390-400 K within the freshly ventilated TTL (BrO > 7.5 pptv) shown during flight SF2 makes me doubt about the ability of the the CH4 < 1790 pptv as a good proxy for distinguishing stratospheric air.

Again it tentatively separates LMS from TTL air, for which unfortunately no strict horizontal barrier or criteria exists to distinguish both compartments.

Also, What is the meaning of showing Fig. 13b., for which most of the panels show no data at all??

Inferred total Br_y^{inorg} in the LMS. Unfortunately, due to operational constraints no measurements are available for Θ < 380 K.

Fig. 14: Flights SF2 and SF4 are not shown. Why not?.

For SF2, GWAS did not measure VSL bromine and for SF4 a DOAS retrieval problem exists (see text).

(P14, L5) It would be a good idea to show the mean results for the campaign, and then highlight results for each of the flights.

Based on our experience we think averaging is often not a good idea. In fact, we provided comparable results once (for IO), which was then incorrectly used (by not properly considering the actual photolysis frequency during the measurements) in follow-on studies.

Fig. 11: I was surprised about the dispersion on the modelled/measured scatter plot for CH3Br. Being the bromo-carbon with largest lifetime, I would expect it to have an equivalent dispersion to the halons. No mention is given about this issue in the text.

Probably your surprise calms considered that the data range from 6 to 7.5 ppt for CH3Br and from 3 to 4 for Halon 1211, i.e. due to the scaling where the point (0,0) is truncated.

Additional Scientific Comments:

P2,L15: Is there a direct assignation of WMO, 2014 values specifically to year 2011?

It is assigned to year 2013, since only publications until 2013 are included in the report.

P3,L1: From which reference/s do you obtain the (2.5-4) ppt error on Bry inorg uncertainties within the inorganic method?

From our own experience (+/-2.5 ppt) and published literature (+/-4 ppt) (c.f., Parella et al., 2013)

P3,L4: UT, TTL and LMS should be defined independently in the abstract and introduction. There is no sense to introduce exactly which definition of the TTL you used in the abstract/introduction.

For the definition of the UT and TTL see the off-line discussion in which you were included. Even more complicated is the separation of the TTL and LMS since there exists no fixed barrier, even not with respect to PV. Since no dynamical variable were measured from aboard the GH, we decided to use the methane >< 1790 pptv criterion to separate both air masses as act of necessity.

P3,L32: Dorf et al., 2008 reported a contribution from VSL of (4.0 ± 2.5) for PG and (5.2 ± 2.5) for total (SG+PG). I do not find where you get the value of (2.5 ± 2.6) ppt. Also, there seems to be a confusion in the interpretation of contributions 3 and 4) from Dorf et al.

We checked our files, and accordingly changed to sentence to "The most direct information on contributions 3 and 4 came by the studies of Dorf et al. (2008), WMO (2011), and Brinckmann et al. (2012). They inferred 1.25± 0.16 ppt (VSLS-SGs, contribution 3) + 4.0±2.5 ppt (PGs, contribution 4) = 5.25±2.5 ppt (contributions 3 and 4) and 2.25±0.24 ppt (VSLS-SGs, contribution 3) + 1.68±2.5 ppt (PGs, contribution 4) = (3.98±2.5) ppt (contributions 3 and 4) from two balloon- borne soundings performed in the TTL and stratosphere over north-eastern Brazil during the dry season in 2005, and 2008, respectively."

P4,L4: Saiz-Lopez et al., 2012 also estimated the climatic impact of VSL sources within a CCM.

We added the reference.

P7,L10: Please describe which species were measured by the GWAS sampler, and which ones were used within this work.

We added a paragraph on P7, L7cont.

P7,L27: "The received limb radiances …". Do you mean the limb radiances measured with the mini-DOAS instrument? Please specify.

We accordingly changed the text to… The measured limb radiances of the mini-DOAS instrument…

P9,L3: Other works used this type of experimental data measurements on top of a model "curtains" (i.e Nicely et al., 2016). Some reference to any of these articles could be given here.

Using predicted curtains in the interpretation retrieval of UV/vis remote sensing data is not new at all. For example, we used it already in 2000 in the Harder et al., study (Figure 1), and later in studies, Weidner et al., 2005, Dorf et al., 2006….. and of course the IR community uses modelled curtains to forward guess their observations since the early 1990s.

P9:L20: What do you mean by "the NASA-ATTREX flights of the Global Hawk were strongly biased with respect to the sampled air masses". Please clarify.

The primary focus of NASA ATTREX mission was to study aerosol and clouds within the TTL (Jensen et al. 2015), and not to probe air of the subtropical LMS. So the flights went from EAFB base straight to the predicted coldest spot in the TTL on the highest possible cruise altitude (also because of air safety reasons, without losing fuel in performing dives in between at daytime). In addition, the GH has only two gears, either full thrust or no thrust on the engine (the latter used to perform the dive)

P11,L27-30: Please specify for which Flights the values for NO2 range between (70-170) ppt and were interpreted as belonging to the LMS.

For the separation of the LMS and TTL see above. The reminder is inferred from the data.

P12,L9: What do you mean by "even if the data is scattered from flight to flight"?

During the flight and from sortie to sortie the measured and modelled source gas data agree fairly well, but not perfectly.

P12,L17: Ordoñez et al. 2012 also describes the geographical and temporal variability of oceanic VSL sources.

….which was not implemented in the present simulation, primarily due to reasons given above (replies to queries 3 and 4). Also the Ordoñez et al. 2012 emission scenario is not undisputed for all VSLS species (see Hossaini et al., 2016).

P14,L6-7: Please explain better what you mean by "very young air" and "older air". Are you considering the [CH4] proxy given before?

We accordingly changed the text to ….. or comparable younger air based on measured [CH4].

P15,L5-7: A printed value of the overall Bryinorg error (and range) derived from the analysis shown on panel f of Fig. 4-9 would be helpful in the text.

Unfortunately, this poses a problem since the error changes from observation to observation due to the factors mentioned in the text. So addressing them in text would require an extended table. Individual errors bars are however given in Figure 14, i.e. for flight section when the WAS instrument sampled air for the brominated organic species.

P15,L22-26: All the analysis about the Bryinorg increment due to the decrease in VSLS is performed based on the "theoretical" dashed diagonal lines shown in Fig. 15. But the "modelled" diagonal lines are not shown for neither of the flights. Why? Including the modelled lines would strengthen the analysis, and justify the conclusions obtained here. Also, what do you mean by "…extrapolating the data points along lines of constant [VSLS]+[Bryinorg] (…) to [Bryinorg] = 0"? Do you mean getting the intercept of each of the (not shown) lines for each flight?

Naively speaking yes. However, the plot expresses something like a 'thought experiment' in that (by incorrectly) assuming that if there were no loss of bromine in the troposphere (which we know is not true), then near surface air would at least need to contain the amount of VSL bromine as indicated by the interpolation to [Bryinorg] = 0. Such a thought experiment is justified based on that (a) near surface VSL bromine measurements are much more frequent than VSLS plus inorganic bromine measurements (the latter virtually do not existing).

P16,L12: It is not clear how the range (0.5 to 5.25) pptv or uncertainty (± 1.04 pptv) of inferred Bry_{inorg} values is computed. Are these the maximum-minimum values modelled with SLIMCAT for all flights? Is this a model average within the Eastern Pacific region where measurements took place?

Yes, the range (0.5 to 5.25) ppt spans the minimum and maximum Bry_{inorg}, and the (2.63± 1.04 pptv) is the average over all inferred Bry_{inorg} at the bottom of TTL (according to our preferred definition). By

the way, the mean amount of inferred Bry_{inorg} compares very well with the model results (at 17 km) mentioned in Navarro et al, 2015 for the Eastern Pacific (i.e. compare Figure 13a the panel for 370 to 380 K results with concentration given in Navarro et al., 2015, Table 1 for the Eastern Pacific, i.e. 5.98 (VSL + Bry_{inorg}) minus 2.96 ppt (VSLS) versus 3.1+/-1.28 ppt (see Figure 13a) of the presented study.

P16,L19: The chemical loss rates were computed within the tropics (i.e, considering the 0º - 360º longitudes) or only within the Eastern Pacific region? It is important to make this clear, and in case of considering the whole tropics, a comparison to the values obtained within the EP is needed.

We re-run the model and accordingly changed first the sentence in the introduction to ….. Finally, for the Eastern Pacific (170_W - 90_W) the TOMCAT/SLIMCAT simulations indicate a net loss of ozone of - 0.3 ppbv/day at the base of the TTL (Θ = 355 K) and a net production of + 1.8 ppbv/day in the upper part (Θ = 383 K)……and second in section 4.7 to….. The chemical rates are averaged over the Eastern Pacific region (20º S - 20 ºN, 170 º W - 90 º W) for the duration of the campaign.

P16,L22-27: The chemical loss analysis gets into details of which independent families and specific channels are the major contributor to the total (or bromine) ozone loss rate. I suggest including a table/sentence defining all the families considered, and which reactions are considered at least for the bromine channel.

We accordingly included an annex explaining which reactions where considered in the ozone loss calculations.

P17,L9: I do not agree that your results provide confidence in the modelled NOy photochemistry. You have only shown results for NO2 in this work. Much deeper analysis of nitrogen cycles should be given in order to perform this statement within the conclusions. Please see also text in P12,L3-4.

We partly concur based on that other NOy species were not measured! However, the NOy species would need extra-long atmospheric life-times since the shorter lived NOy species (N2O5, HO2NO2, HONO, ClONO2, BrONO2) are readily photolysed in the TTL at daytime, and would add to NOx, and ultimately to NO2 (being in steady state with NO). Even a longer lived NOy species, such as HNO3 need to be correctly modelled, because its life-time is certainly shorter than that of CH4. If not true, it would it again add to NOx.

Technical/Linguistic Comments:

P2,L11: It is not necessary to include a minus sign whenever you state that the value represent a net destruction. There are many other places in the MS when this also occurs.

Yes but nevertheless correct.

P2,L15: Within the text and figures, the terms Bry and Bry_{Total} are both used to represent the same quantity. Please unify the criterion.

We changed Bry^{Total} to Bry where necessary (cf. Figure 14).

P2,L17. All halons must be written either with capital H or lower case h.

We changed all halon to Halon.

P2,L32: … several tenths of ppt.

We accordingly changed the text.

P3,L15: Colombia, not Columbia

We accordingly changed the text.

P5,L29: UAS acronym is not used at all in the paper, there is no sense to define it. Also, if the size is in Liters, then capital L should be used.

We accordingly changed the text.

P13, L3-7: Consider rephrasing.

We changed to text to…. Second, even though Wang et al. (2015) used a technique similar to ours and in particular they use the same radiative transfer code (e.g., McArtim see Deutschmann et al. (2011)), they inferred the BrO profiles using the optimal estimation technique (Volkamer et al., 2015).

P14,L17: What about BrCl? It is shown in Figs. 4-9 but not included in the definition of Bryinorg?

We accordingly changed the text.

P14,L19-20: Have you thought on including in a table the most important reactions that were changed between the three sensitivity runs?

No since the reaction are well known and are already tabulated in the JPL compilations.

P15,L35: Is the call to Fig. 15. correct?. If so, please explain.
Yes it is correct.
P17,L6: do you mean anti-correlated?
We accordingly changed the text.
Fig. 2: The altitude range should be given in between brackets.
We accordingly changed the text.
Fig. 15: Consider rephrasing the sentence starting with: “If all Bry …” for one in the form “The dashed diagonal lines indicate …”

We changed the text to “If all Bryinorg resulted from destroyed VSL bromine of the same air mass from near the surface, then all data points should follow individual diagonal lines.”

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