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.

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.

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.

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 Bry^{inorg}, the authors should describe specifically how the model was constrained or adjusted. For example:
    a. 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 Bry^{inorg} 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.

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?

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.
    a. 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.
    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 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 Bry^{total} should also be discussed.

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.

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). 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. 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 surpass 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?.

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. In P11,L17-20 you state that "The excellent

agreement achieved between measured and modeled 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.

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.”
   P4,L1: “… indicating that some Bry_inorg (i.e. several ppt) is directly transported from the tropopause”
   P9,L17: “…, some exciting observations and details”
   P11,L4: “… we refrain from this much more complicated approach”
   P11,L27: “… NO2 meet the expectations for NOx”
   P16,L24: “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.
   P12,L3: NOy=(NOx,N2O5,HNO3,HO2NO2,…. What the three dots means? Please specify.
   P17,L14: Once again [CHClBr2,CHCl2Br,CH2ClBr,…]
   P18,L3: (Here: cite Hossaini 2016, acp when published)?

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?

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.

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

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

Fig. 14: Flights SF2 and SF4 are not shown. Why not?. It would be a good idea to show the mean results for the campaign, and then highlight results for each of the flights.

Fig. 11: I was surprised about the dispersion on the modeled/measured scatter plot for CH3Br. Being the bromocarbon 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.

Additional Scientific Comments:

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

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

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.

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.

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

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

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

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.

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.

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.

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

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

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

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.

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?

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?

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.

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.

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.

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.

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.

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

P2,L32: … several tenths of ppt.

P3,L15: Colombia, not Columbia

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.

P13, L3-7: Consider rephrasing.

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

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

P15,L35: Is the call to Fig. 15. correct?. If so, please explain.

P17,L6: do you mean anti-correlated?

Fig. 2: The altitude range should be given in between brackets.

Fig. 15: Consider rephrasing the sentence starting with: “If all Bry …” for one in the form “The dashed diagonal lines indicate …”