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Interactive comment on “Modelling the Inorganic Bromine Partitioning in the Tropical Tropopause over the Pacific Ocean” *by* Maria A. Navarro et al.

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Responses have been uploaded also as a supplement pdf file. Please refer to this file as the "plain text" option do not show equations or reactions added to the responses.

Review of: Modelling the Inorganic Bromine Partitioning in the Tropical Tropopause over the Pacific Ocean by Navarro et al.

The manuscript of Navarro et al., reports on Cam-Chem (Community Atmosphere Model with Chemistry) modelling of the partitioning of inorganic bromine in the tropical tropopause layer(TTL) over the eastern and western Pacific Ocean. The modelling is compared with (averaged) observations of some key species, i.e. of the in-situ measured brominated source gases and O3 from which to the partitioning of inorganic bromine is concluded. Comparisons of measured with and modelling in particular for

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Discussion paper

the yet underexplored TTL are per-se important and interesting. However, based the already-published literature and state knowledge of this field, the paper has major flaw in its present state. My criticism of the present study is based on 5 major deficits (2 more general and 3 more specific comments including one related remark, #3), which are detailed in the following:

Response: We thank Prof. Pfeilsticker for his comments. We really appreciate and welcome any feedback that could be used to improve our manuscript.

I.) Methodological deficits of the study 1. Using (spatial and temporal) averages for fast reacting species (radicals) in photochemical calculations: For the modelling of the bromine partitioning, averaged in-situ O3 (together with bromine released from brominated VLSL) is used. Averaging over space and time of concentrations of photochemical reactive species (Figure 1) however is dangerous, since it may lead to incorrect and spurious results for the inferred quantities (for example concentration ratios). In order to see this please consider the rapidly established steady state of [Br] and [BrO] (both being a function of space x and time t) as well as of some (radical) species (e.g. O3, OH, HO2,..) at daytime, which is established through eq. (1) (see eq in pdf version) where in the present context irrelevant and missing terms are abbreviated by …. . Evidently in order for the equation to make sense k(T), O3(x,t), JBrO(x,t) need to be local (i.e. measured or calculated) quantities in the photochemical calculations. When using instead space and/or time averaged quantities (the overbars denote either space or time averaging), the above mentioned equation would instead read as eq. (2) (see eq in pdf version) It can easily be seen, however, that the [BrO]/[Br] ratio calculated from averaged (space or time) quantities and from local quantities generally differ eq. (3) (see eq in pdf version) and accordingly only the latter gives the right answer for the photochemically established [BrO]/[Br] ratios in the atmosphere. In conclusion, when using space and time-averaged ozone concentrations (from the manuscript it is not clear as to whether (k(T), JBrO, [Br] and [BrO] were also averaged in the same manner or not, but the answer is somewhat irrelevant to my argument), the modelled [BrO]/[Br] ratio may depart more or less from the actual atmospheric [BrO]/[Br] ratio.

Response: We agree with the reviewer regards the different results eq. (1) and (2) can give depending on the spatial or temporal average applied to either modelled and/or measured data. In doing so, please note the importance of assuming that the "irrelevant and missing terms" for the case of the [Br]/[BrO] ratio are negligible compared to the dominant production and loss channels for atomic bromine in the atmosphere. Only if those irrelevant terms can be neglected, then eq. (1) and (2) can be written in its simple ratio form (i.e., dependent only on the photodissociation rate constant JBrO and on the pseudo first-order reaction rate k(T)*[O3]). Having said this, we would like to make the following points clear: - Photochemical production of bromine atoms in the Upper Troposphere (UT) is dominated by BrO photolysis (see reaction in pdf version), while chemical losses occur mainly through the bimolecular thermal reaction (see reaction in pdf version), which itself represent 98% of the total atomic Br losses, (see Saiz-Lopez and Fernandez, GRL, (2016) for details). The much rapid reactivity of these two channels (respect to the neglected terms in eq. (1)) allows the establishment of a rapid pseudo steady-state between Br and BrO. Thus, if we accept that a rapid steady-state is reached between these two species, and also agree that neglected terms are irrelevant for this case, then it is evident that Br and BrO abundances must be related by a mathematical expression which considers only the reaction rates connecting those species. - We do not mention at all the explicit relation between the [Br]/[BrO] ratio and JBrO, k(T) and O3 in this manuscript. We only mentioned the relevance of computing the [Br]/[BrO] ratio in relation to the proposed tropical rings of atomic halogens, whose drivers are described in a preceding paper (Saiz-Lopez and Fernandez, GRL, 2016). Anyhow, and being aware of the averaging issues mentioned by the reviewer, in that work we performed spatial and temporal averages of Br and BrO abundances, as well as to all rate constants affecting atomic bromine production and losses, and found an excellent correlation between instantaneous (e.g., hourly) and averaged (e.g., monthly) modelled output. Indeed, Fig. 3 in Saiz-Lopez and Fernandez, GRL, (2016), shows the vertical profile of the [Br]/[BrO] ratio as well as the JBrO/K(t)[O3] ratio obtained with CAM-Chem for an equivalent setup simulation as the one used in this work. The main panels show either annual values for the tropical (20°N-20°S) average (Fig.3D) or monthly values within the Tropical Western Pacific (TWP, Fig.3E), while the inset panels show the hourly output modelled linear correlation (daytime masked) between[Br]/[BrO] and JBrO/K(t)[O3]. For the case of the Tropical region, r2 = 0.9782, while for the TWP, r2 = 0.99695, with ratio values spanning approximately from 0 to 3. Equivalent results were also obtained when individual model gridbox (lat,lon,z) were sampled, either hourly or monthly. - The model output for the present simulations was instantaneous (i.e. hourly, half-hour could have been the highest possible resolution within CAM-Chem). The model was run in Specified Dynamic (SD) mode (i.e., considering the current meteorology prevailing during the campaign) and was further sampled at the correspondent latitude, longitude, height and time “gridbox(lat,lon,z,t)” that best matched the ATTREX flight-track. Thus, nor spatial neither temporal averaging of [Br], [BrO] and its ratio [Br]/[BrO] or other atmospheric quantities in CAM-Chem (JBrO, K(T), T, O3, etc.) have been performed to extract the model output. As by the time of preparing this MS there were no other measurements available than ozone, we decided to present the validation of instantaneous O3 measurements for all independent flights (Figure 1, now Fig. 2 in the revised version) and then all atmospheric model variables were averaged into 1 km height bins so the output correspondent to each independent flight could be compared with each other. Being this one a modelling paper (as it is clearly stated in the title), we found appropriate to also perform the spatial-temporal mean of all flight-tracks, which are then used to present a more general representation of the modelled state of the atmosphere in the rather yet unexplored tropical upper troposphere (e.g. the Vertical Profiles shown in Figs. 4, 7 and 8). We are aware that these mean vertical profiles are not descriptive of each of the independent flights, but they are certainly representative and illustrative of the mean state of the tropical atmosphere within the Eastern and Western Pacific when sun photochemistry is turned on and off. - For the case of Fig. 5, which shows the temporal evolution (i.e., SZA dependent) of the dominant bromine species and the main inorganic reactants during the day, twilight

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and night, we decided based on Prof. Pfeilsticker′s comment, to present in addition to the mean temporal profile, the independent results for each specific flight. In this way, the changes in partitioning of the dominant species can be addressed directly in response to the current abundance of ozone, Cly and/or NO2 prevailing during each flight track. This helps, for example, to support the large inhomogeneity we suggested in previous studies (see Saiz- Lopez and Fernandez, GRL, (2016) for details) for the [Br]/[BrO] ratio, which is modelled to be larger than 1 at a fixed SZA for one of the flights but not for the others: for this cases, as highlighted by the reviewer, performing the spatial-temporal mean of all flights does not illustrate the intrinsic variability found on the abundance of ozone, bromine and all related short-lived quantities. Following the Prof. Pfeilsticker′s comments and the above responses, we have modified the text in as follows: Page 5 line 20: “Model hourly output was sampled at exactly the same times and locations as the ATTREX measurements, without performing neither spatial nor temporal averaging on model grids. Once each independent flight track was extracted from the model output, all atmospheric quantities were averaged into 1 km altitude bins, to compare with measured data.” Page 7 line 23: “Figure 5 compares the mean abundances observed in the EP and WP considering all flights. Even when these results are not descriptive of each of the independent flight, they are representative and illustrative of the mean state of the tropical upper atmosphere within the eastern and western Pacific in the presence and absence of sunlight. Equivalent results but for each independent flight are show in the Supplementary online material” Page 9 line 1: “A closer inspection on each independent flight (Figs. S1 and S2) reveals the large inhomogeneity of the tropical rings of atomic bromine. In the EP, Br surpass BrO mixing ratios at 60° SZA for flights RF04 and RF06, but as the remaining flights sampled larger BrO mixing ratios, the mean EP abundances shown in Fig. 5c shows Br/BrO > 1 only at 20° SZA. Similarly, the mean results shown in Fig. 5a for the WP show BrO > Br at all times, but RF02 and RF03 show the ratio Br/BrO to be larger than one at 50° SZA. This highlight the importance of considering non-averaged (both spatially and temporal) model output to determine the concentration of photochemical reactive species or other atmospheric quantities such as the Br/BrO ratio." Page 9 line 14: "Figure 7 shows the distribution of the Br/BrO ratio over the WP and EP, and its correlation with ozone concentrations and temperatures. The results are based on the mean 1 km binned data for all track flights, although equivalent conclusions can be reached for each independent transect."

Further when inspecting the ozone concentrations measured by the NOAA instrument in the TTL during ATTREX, it can be seen that actual ozone concentrations may vary by up to a factor of 10 (mostly with height, less in the horizontal in the TLL, see Figure 1) and so the [BrO]/[Br] should cover a similar dynamical range (keeping all the other parameters the same, see Figure 8 in Fernandez et al., (2014)), a behaviour not really recovered when using 1 km binned averages for ozone (Figure 1). As consequence, the modelled [BrO]/[Br] may not well represent actual [BrO]/[Br] ratio met in the atmosphere, and as thus may not really provide a meaningful information to reader.

Response: The sensitivity of the [Br] and [BrO] abundances to ozone mixing ratio shown in Fig. 8 of Fernandez et al., (2014), was performed using a box-model constrained with many chemical parameters (not relevant to described here) and also constant temperature (T=190 K). Note that Fig. 9 of the same paper, shows an additional sensitivity of bromine abundances to temperature. As the bi-molecular thermal reaction (Br + O3—> Br, see equation in PDF version) decrease with increasing temperature (K(T)= 1.60 x 10-11 e(-780/T)), we do not expect the modelled [Br]/[BrO] ratio to cover a similar dynamical range than the ozone variations: it would also depend on the temperature change associated to the air parcels considered (K(T) changes a factor ∼ 1.5 between 190 and 210 K). Also note that in the current modelling approach, ATTREX ozone measurements has been used to validate CAM-Chem performance (Fig. 1), but individual measurements values have not been used to compute the [Br]/[BrO] ratio. The modelled [Br]/[BrO] ratio shown in Fig. 7A was computed considering CAM-Chem ozone (Fig. 7B) and temperature (Fig. 7C) fields. Modelled ozone abundances change between 100 ppb and 600 ppb and [Br]/[BrO] ratios between 0.35 and 2.0, so the modeled range between maximum and minimum values span approximately a factor of 6. With regards to the last sentence, in any case, spatial/temporal average of the modelled [Br]/[BrO] ratio may depart more or less from the actual modelled JBrO/K(T)[O3] ratio, but not such a strong affirmation can be made respect to the atmospheric [Br]/[BrO]. There are no means we can compare here modelled [Br]/[BrO] with atmospheric [Br]/[BrO] ratios as that would have implied the simultaneous atmospheric measurements of atomic Br and BrO. Even when Br atoms (as well as atmospheric Bry) can be inferred from BrO measurements, this procedure also implies including a detailed chemical mechanism for bromine. Thus, any modelled ratio (such as [Br]/[BrO], [Br]/[Bry] or the more commonly used [BrO]/[Bry] partitioning) can be compared to atmospheric ratios as long as the chemical mechanism considered is appropriate to represent the chemistry of that specific portion of the atmosphere. We are quite confident that bromine chemistry in CAM-Chem is very well represented (as in many other global models) and that all the main chemical reactions reported in the literature are up-to-date in our setup. Thus, we found quite interesting to compute atmospheric ratios between the major species to establish which ones are the dominant species, and in this way, validate them against measurements to properly constrain chemistry-climate models.

Finally, the ozone measured by the NOAA instrument and plotted in Figure 1 (right panel) appears to be spikier (due to any reason, but this could also be visual illusion) than the same ozone plotted in Figures 3 to 8 (panel c) in Werner et al., (2017) for the Eastern Pacific.

Response: Measured ozone values have been processed as described in Section 2.1. We found quite difficult to compare the Vertical Profiles shown here in Figure 2 (note that Fig. 1 was shifted to Fig. 2 in the revised manuscript) with the temporal timeseries shown for each flight in Werner et al., 2017, Figs. 3-8c. In any case, the spiker representation of O3 measurements presented here might explain the large range between the maximum and minimum [Br]/[BrO] ratios you expected to find.

2. Averaging (over the space and time domains) concentrations for longer lived species: For some selected measurements (which ones?) 6 O3 averages and corresponding averages of CHBr3, and CH2Br2 (out of in total 745 in-situ samples from the EP according to the information provided in Figures 2 and 3a) are inter-compared with the respective model predicted parameters. Averaging over time (or space) for species of different photochemical lifetimes is somewhat problematic. In order to see this let's consider species of different photochemical lifetimes τi (i = 1, 2, 3...) with a common timescale against atmospheric transport τm. Here remember that in general photochemical and dynamical time scales for individual air masses are distributed in space and time (e.g., Waugh and Hall, 2002; Waugh, 2009; for TTL distributions of O3 see c.f., Pan et al., 2014). For the moment, however I skip these complications. Then the joint timescale for photochemical processing and transport is given by where for the sake simplicity, it is assumed that both photochemical and dynamical processes lead to exponentially decaying concentrations. With these simplifications in mind, the time averaged concentration is then obtained from (which is a Laplace transform of c(x,t)). Averaging samples using an appropriate kernel (here exp(- t/teff, i) is of course different from the (geometrical) average taken over individual samples of ck(x,t), i.e. since in the latter calculation any kernel (whether appropriate, or not see below) to calculated averages is discarded. While for photochemical processes an exponential decay is a reasonable assumption, for dynamical processes in the atmosphere, it is certainly not a good assumption due to the turbulent transport (2-D in the stratosphere). Accordingly, the kernel for dynamical averages (often also called probability density functions, or pdf) does not follow an exponential but rather a power law (e.g., Min et al., 1996; Pierrehumbert and Yang, 1993; Minschwaner et al., 1996; Seo and Bowman, 2000; and for the statistics of actual field data of O3, ClO, and others e.g., see Tuck et al., 2003; Tuck, 2008; Pan et al., 2014). As a consequence, the resulting air mass age spectrum (from which the average age can approximately calculated) is then (approximately) represented by Γ-type functions for the concentrations, which again depend on the time and location in the atmosphere (Hall and Plumb, 1994, Waugh and

Hall, 2002). In consequence, the comparison of modelled and measured averages (for ozone in Figure 1 and 2, and ozone CHBr3 and CH2Br2 shown in Figure 2) does not really make sense, if the pdfs for the atmospheric and modelled samples are not the same in a statistical sense. To put it into simple terms, when averaging over (limited) samples one has to prove that the sampling from the real atmosphere and from the modelled atmosphere are made from the same statistical distributed event in space-time manifold in order for comparisons to make any sense. So certainly the way that the measured and modelled parameters are averaged deserves much more attention in the manuscript. Finally, noteworthy is that averages over temporally and spatially distributed 'fluctuations' only give the same result for the inferred moments (averages, variance, et cetera) if the system is ergodic, which unfortunately in atmospheric dynamics is mostly not the case. Moreover, the samples need to be huge in order to fulfil one requirement of the central limiting theorem (CLT), that both samples (taken from the atmosphere and the model) converge to the same pdf (given they are the same which needs separately to be proven).

Response: We appreciate the explanation but we believe that the concerns found by the reviewer in this point are not relevant for this study.

3. Comparing remotely sensed and modelled concentrations: Moreover, the kernels to calculate averages (and used further on in inter-comparison exercises, see below) in remote sensing applications and in inverse modelling are strongly instrument and measurement-dependent (Rodgers, 2000). Fortunately, they often mask the above described effects due to their limited spatial or time resolution, i.e. their inherent averaging. In fact, in the latter applications these 'kernels' are called 'averaging kernels (AK)' of the observation and in colloquial English the averaging kernels can be called the 'glasses' by/through which the remote sensing observations were made. So the characteristics of 'the glasses' need to be considered in some way in inter-comparison exercises with modelled quantities (see below). For some examples of actual AKs, please inspect Figures 5 and 10 (for the weighting of the probed concentrations in the

horizontal) in Stutz et al., (2016), Figure 3.5 in Rodgers (2000), Figure 1 (below), or any other study on remote sensing. Chapter 3 in Rodgers, (2000) also discusses the different error sources of the traditional inversion methods used in remote sensing and inverse modelling. It also describes how remotely sensed quantities (here called co(i), where i is the retrieval grid number somehow representing the vertical resolution of the measurement) need to be compared with modelled results (cm(i)), i.e. by comparing the inferred co(i) with the product AK· cm(i), where AK is a tensor, of which the columns (or rows) a filled with the individual averaging kernels, displayed for example in Figure 1. In order to avoid these complications using traditional inversion methods for the interpretation of remotely sensed quantities (and in the particular case those arising from multiple scattering due to the a priori unknown spatial distribution and optical properties of aerosol and cloud particles), Stutz et al., (2016) describes a novel (scaling) method for the interpretation data. In effect, the scaling method uses additional information gained from simultaneously in-situ measured gases (i.e. O3) in order to assist the interpretation of remotely sensed NO2, and BrO in the TTL. Therefore, the scaling method has to be considered as a hybrid method (since it uses information collected by remote sensing and in-situ measurement), which comes with some advantages (and disadvantages) over traditional remote sensing methods. For example, it provides a higher accuracy than methods purely relying on remotely sensed information. Evidently the major disadvantage of the scaling method arises from the need of in-situ information of the probed air masses, i.e. it is suitable for applications from satellites, or high flying balloons. Further the scaling method still requires to carefully consider (by RT calculations simulating the observations) in order simulate how the information (the measured absorption) is obtained. Accordingly, when applying the scaling technique to their remotely sensed data, Stutz et al., (2016) and Werner et al., (2017) actually simulated each individual observation by modelling the actual RT (and the predicted absorption of the targeted species) by considering instrumental and other details of the measurements as well as predicted curtains of the targeted species, obtained from CTM modelling (TOMCAT/SLIMCAT). This approach (as in any traditional remote sensing application) thus carries over to the analysis any relevant instrumental and observation-related features in the forward modelling of the observation. Evidently, the scaling method (as any traditional inversion method) then allows very close intercomparisons of the predicted quantities (e.g. trace gas concentrations) with the observations, including a correct attribution of the fraction of the measured absorption (or slant column) to parts of the atmosphere not directly probed by the observation, however only if the averaging kernels are appropriately considered. Here please also note that the latter approach to inter-compare remotely sensed data and CTM modelling is not new at all, but e.g., it has been used by our group for more than 2 decades. Further using the scaling method, the calculation of absolute concentrations is achieved using a simultaneously in-situ measured and remotely sensed gas (e.g., O3), together with an appropriate consideration (by RT modelling) of the different sensitivities for detection of the targeted and scaling gas (see equation 14 in Stutz et al., 2016). In effect, the accuracy of the inferred quantities is arguably much better (Stutz et al., 2016) than only relying on remotely sensed quantities for the retrieval of concentrations. Accordingly check your statement on page 6 (lines 14 and 15) for correctness.

Response: we thank the valuable information the reviewer gives us about remote sensing techniques and the scaling method, but our manuscripts is not based on them. We are not using the CAM-Chem model to compare with any remote sensing data. The manuscript is based on the discrete measurements taken with in situs GWAS, which CAM-Chem reproduced very well.

II.) Comparison with available measured data Further, I'm really curious why the authors did not attempt to compare their modelling work with actual measured NO2 and BrO data (potentially) available to the first author for more than a year and which now have been published (Werner et al, 2017). However, when using remotely sensed data in inter-comparison exercises, the kernel for horizontal averaging (see Figure 10 in Stutz et al. (2016)) has to be appropriately taken into account for the modelled data (see my remark #3 above). Further, given that the Werner et al., (2017) manuscript (which the first author of the present article co-authored) was submitted earlier (July 17, 2016) than the present manuscript (Nov. 18, 2016), the statement on page 4 (line 18) is not well based. By being more specific, the lack of a tight comparison of the modelled results with existing measured data give rise to some more deficits of the present study: 4. Simulated NO2: For the Eastern Pacific TTL, the CAM-Chem model predicts NO2 between 0.7 – 343 ppt at daytime (Table 1 and Figure 6). No reasons are provided for the elevated NO2 in the TTL over the EP, except that modelled air masses are affected by ‘pollution’. However, no other indication (neither from, for example, measured CO during NASA-ATTREX (UCATS) nor any further evidence inferred from the model) is provided that in fact polluted air masses were reaching the TTL over the EP in early 2013. In fact, the NO2 mixing ratios reported by Werner et al., (2017) were < 20 ppt in the TTL, and they agree well (within the error bars +-/10 ppt) with the predictions of the TOMCAT/SLIMCAT simulations assuming no contribution from ‘pollution’. In all these respects, and in particular with respect to the discussion provided above under point #3, the statement on page 6 (lines 14 and 15) is not well founded. Accordingly, in any further study information has to be provided why for the EP TTL the Cam- Chem model predicts NO2 concentrations much large than observed. In addition, coherent evidences both from observations and modelling has to be provided (for example from CO/O3 and CH4/ O3) that indeed the TTL over the EP is affected by ‘pollution’. 5. Simulated inorganic bromine, its partitioning and spatial patchiness: A major part of the study is devoted to model the bromine partitioning. First, I found it hard understand why the model does not really reproduce the increase in total inorganic bromine with increasing height (potential temperature) within the TTL, mainly caused by the destruction of brominated VSLS. This is somehow curious since the bromine concentrations at the lower boundary reported by Navarro et al., (2015) (page 3, line 18 and 19; VSLS; 3.84 $\pm$ 0.64 and 3.18 $\pm$ 1.49 ppt from WP and EP, respectively, and inorganic bromine 3.02 $\pm$ 1.90 ppt of Bry over the EP and 1.97 $\pm$ 0.21 ppt over WP) are in reasonable agreement with the data for the EP TTL, reported by Werner et al., (2017). Moreover, in the Cam-Chem model inorganic bromine (in gaseous form) barely increase from ∼2 ppt (from the lower boundary at 14 km) to ∼3 ppt at 18 km (Figures 4 and 8), in stark contrast with the observations presented in the Werner et al., study for the upper levels of the TTL over EP. Here, depending on the flight, inorganic bromine ranges from (2.63 ± 1.04) ppt (range from 0.5 ppt to 5.25 ppt) to 5.1 ±1.57 ppt (at Θ = 390 - 400 K) to 6.74±1.79 ppt (at Θ > 400 K), in agreement with the measured destruction of brominated VSLS species (Navarro et al., 2015, and Figure 14 in Werner et al., 2017). So the obvious question is: Does the model either not efficiently destroy the brominated VSLS, and/or does the missing bromine reside in/on particles? If the latter is the case, the bromine up-taken by particles need to be rather large (2 – 3 ppt) in order close the bromine budget. So some information has to be provided how the bromine budget is closed in the model, and in particular on how much bromine is up-taken by the particles. Next even though the modelled absolute amount of gaseous inorganic bromine likely may not affect the Bry partitioning, the modelled [Br]/[BrO] (cited: (1) . . .. the modelled Br/BrO maximizes at 17 km from page 7, line 7 to 17 and in Figure 7 and (2) . . ...that Br/BrO may become as large as 2 in the TTL of the EP, see Figure 7) deviates from expectations based on the amount of ozone and its increase with height (see Figure 1 left panel, and Figure 3 to 8 in Werner et al., 2017), and the modelled bromine partitioning in the TTL as function of ozone (Fernandez et al., 2014, Figure 8). In fact, these findings largely contrast with early findings based on the Br/BrO ratio in TTL (at 17 km) during daytime c.f., by Fernandez et al., 2014 (Figure 1, left panel where Br/BrO < 0.6 at 17 km during tropical noon), Schmidt et al., (2016) (Figure 1), or lately the model results presented in Werner et al., (2017) (inspect Figure 3 – 8, Br/BrO < 0.6 at 17 km). Reasons for this discrepancy, including a discussion how the averaging of the ozone and the source gas concentrations and of other quantities impacts the modelled Br/BrO ratio (see points 1 and 2 above) certainly need to be addressed in any future study. Finally, the model predicts a certain patchiness (on spatial scales of some hundred kilometres) of the modelled Br/BrO ratio at 17 km for the EP (and WP), with [Br]/[BrO] ratios ranging from below < 0.5 to about 2. No further reason for this patchiness is provided in the manuscript. If air masses entrained by mesoscale convection

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Discussion paper

into the TTL are responsible for this patchiness, then it also needs to be seen in other gases (e.g. CO, CH4. . .), but again no evidence for this is provided in the manuscript. The predicted patchiness also contrasts with measured O3, NO2, and BrO, in particular since the remote sensing measurements can easily resolve horizontal variations of the measured quantities on the hundreds of kilometre scale (e.g., Stutz et al., (2016) figure 9, and Werner et al., (2017), figures 3 to 8). Further, since at daytime a rapid steady state is established between Br and BrO (see above) as function of the solar illumination and O3 concentration, it is difficult to infer from measured data any reason for the predicted patchiness in the [Br]/[BrO] ratio.

Response: As answered to reviewer 2 (see major comment 1) the manuscript has been modified to emphasize the fact that this model study was performed simultaneously to the study published by Navarro et al., 2015. We prefer not to go into further details about the issue of data availability and, although we really thank Prof. Pfeilsticker for his insightful comments, we would also like to think that the soundness of our modelling paper is not solely based on how well or not we compare to measurements and TOMCAT/SLIMCAT model results in Werner et al., 2017, as it seems to transpire throughout this review. Nevertheless, and when relevant, we have also modified the manuscript to state how our results compare to the work of Wener et al., 2017. A final point, so far and to the best of our knowledge, the BrO and NO2 measurements from ATTREX 2014 (Western Pacific) are still being reviewed. Werner et al., 2017 report measurements of BrO and NO2 from ATTREX 2013, although that paper was still in discussion by the time of our submission.

6. Error and uncertainties: Finally, as an experimentalist who devotes 85 % of his efforts in the interpretation of data to get a handle on a reliable (thus justifiable) errors and uncertainties of the measured quantities, I always find it curious if studies lack a proper discussion of errors and uncertainties of the presented results. In modelling studies, this could for example be done by (1) inspecting respective Jacobians of the relevant quantities, (2) investigate differences in the modelled fields from ‘on and off’

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Discussion paper

runs, and (c) perform ensemble runs et cetera. So also in this respect, the present study largely lacks this requirement for robust science.

Response: This is a good point - in this work, we are using the model and chemical mechanism employed in the Navarro et al., 2015 paper. This mechanism was already tested, tuned and validated for tropical vertical profiles of speciated Bry (Fernandez et al., 2014; Ordoñez et al., 2012). The details about the development of the chemical mechanism, all sensitivity tests performed to tune the model along with uncertainties estimation can be found in those previous works.

Summary Given the above described methodological deficits (points 1 and 2), the lacking comparison of the modelled results with actual measured data (points 4 and 5), and the lacking discussion of errors and uncertainties (point 6), unfortunately it is impossible to recommend the manuscript for publication in the present form.

Response: We have addressed point-by-point the reviewer's comments relevant to this work and we appreciate those other clarifications about comparing remote sensing data to an atmospheric 3D model, which are out of the scope of this paper.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/acp-2016-1031/acp-2016-1031-AC1-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-1031, 2016.