We would like to thank the reviewers for their comments which have improved the manuscript. Specific comments from the reviewers are given in bold below, followed by our responses in plain text.

Referee #1

Stone et al. presents a study on the influence of tropospheric halogens on OH and HO_2. The study is based on observations from the Cape Verde Atmospheric Observatory, a chemistry box model, and a global chemical transport model. Overall, the paper is well written and well suited for ACP. I recommend it being accepted following minor revision. I list specific comments and questions below:

1) Line 49: Le Breton et al (2017, doi: 10.1016/j.atmosenv.2017.02.003) have also presented measurements of BrO in the MBL, consider including a citation.

We have included the citation.

2) Line 224: What are the processes behind "Physical loss", is it deposition? If so, I suggest you state this explicitly. Is a lifetime of 24 hour reasonable?

The physical losses incorporate losses such as wet and dry deposition and diffusion out of the box for any model generated species with a sufficiently long lifetime. In our previous work (Stone et al., 2010; Stone et al., 2014) we investigated the sensitivity to the deposition rate, with results indicating that variation of the deposition lifetime between 1 hour and 5 days results in little change to the modelled concentrations of OH and HO_2. We have added the following details to the manuscript for clarification:

Deposition processes, including dry deposition and wet deposition, and diffusion are represented in the model by a first-order loss process, with the first-order rate coefficient equivalent to a lifetime of approximately 24 hours. As discussed by Stone et al. (2010), variation of the deposition lifetime between 1 hour and 5 days results in limited changes to the modelled concentrations of OH and HO_2.

3) Line 244 to 253: The assumption of constant BrO and IO between 9:30 and 18:30 seems overly crude. As can be seen in Figure 3 of Read et al. (reference given in manuscript), it takes approximately 6 hours for BrO and IO to raise from 0 to max. How would the results of the box model study change is more realistic assumptions about the diurnal cycle of BrO and IO are used?

The peak daytime concentrations of BrO and IO are typically reached between 9 am and 10 am, with increases from zero occurring from approximately 6 am, as shown in Figure 3 of Read et al. The discussion in the manuscript is unaffected by the early morning concentrations of BrO and IO owing to the focus on the midday (11 am to 1 pm) concentrations of OH and HO_2, and the HO_2:OH ratio, which are influenced on short timescales. The data presented in Figures 1-3 are potentially affected by the assumption of zero concentrations of BrO and IO before 9:30 am, and we have performed simulations in which concentrations of BrO and IO are set to increase from 6 am until 10 am, at which point the constant concentrations are reached, using the data shown in Figure 3 of Read et al. However, the low concentrations of BrO and IO during this period, and the relatively few data points for OH and HO_2 during these times (there were no measurements of OH or HO_2 at these times of day during SOS2, as shown in Figures 1 and 3), the data presented in the manuscript are not significantly affected. This is shown in the figure below (Figure S6), combined with the response to referee 2. We propose to combine this figure and to provide the details in the supplementary information to the manuscript.

4) Line 288 to 291: Please specify the additional bromine reactions that were added to the Parella mechanism.

We apologise for the confusion here. The additional bromine reactions are those described by Parella et al. (i.e. in addition to the standard mechanism which does not include any halogen chemistry). We have removed the reference to "additional" in order to clarify this:

Emission rates and bromine chemistry included in the model are described in detail by Parella et al. (2012), with the bromine chemistry scheme described by 19 bimolecular reactions, 2 three-body reactions and 2 heterogeneous reactions using rate coefficients, heterogeneous reaction coefficients and photolysis cross-sections recommended by Sander et al. (2011).

5) Line 477-481: Consider extending the discussion of sea-salt debromination. Schmidt et al (reference given in manuscript) presented two simulations: one without sea salt debromination and one including it. The simulation that included sea salt debromination led to levels of BrO that appeared biased high compared to satellite observations, but reproduce the levels observed in the mid Atlantic MBL (see Figure S8 of Schmidt et al.). How would the result of the global model change is sea salt debromination was included?

We have expanded the discussion of this in the manuscript to include the following details:

The global model simulations reported here predict average mixing ratios of ~0.5 ppt for BrO and ~1 ppt for IO during SOS, and thus underpredict BrO but perform well for IO. The underprediction of BrO at Cape Verde results from recent model updates which exclude emissions of bromine species from sea-salt debromination (Schmidt et al., 2016) in order to provide improved agreement with observations of BrO made by the GOME-2 satellite (Theys et al., 2011) and in the free troposphere and the tropical Eastern Pacific MBL (Gomez Martin et al., 2013; Volkamer et al., 2015; Wang et al., 2015). If sea-salt debromination were included, daytime mixing ratios of BrO at Cape Verde would be approximately 2 ppt, as shown by Parella et al. (2012) and Schmidt et al. (2016), and thus in closer agreement to the observations. Increased modelled concentrations of BrO at Cape Verde resulting from inclusion of sea-salt debromination would have a greater effect on OH and HO_2, leading to more significant decreases in OH and HO_2 when bromine chemistry is included without iodine chemistry, with the larger decrease in OH potentially off-setting the increase in OH observed when bromine and iodine chemistry are combined. However, the current model simulations do not consider the coupling between bromine and sulfur chemistry, which may represent a significant sink for reactive bromine species in the troposphere and balance sources from sea-salt debromination (Chen et al., 2017). These results thus demonstrate the need for further investigation and constraint of sources and emission rates of bromine species, and of the coupling between sulfur chemistry and reactive bromine species. We now discuss the global impacts of halogen chemistry.

Also, consider commenting on the resent study by Chen et al (2017, doi: 10.1002/2017GL073812) that show that sulfur chemistry may provide a missing sink of Bry in the MBL to balance the sea salt debromination source.

We have now included details of this study in our discussion of sea-salt debromination (see above).

Referee #2

The paper presents an interesting comparison between observations of OH and HO_2 at the Cape Verde Observatory and two modelling approaches to evaluate how bromine and iodine chemistry impacts HO_x. The results obtained by the two different model setups are discussed and evaluated in the context of the different chemical schemes and timescales considered in the two models. I found the paper interesting and well structured, presenting results in a clear and complete format and hence I recommend the paper for publication in ACP. Please find below two questions and minor comments for consideration by the authors:

- In page 9, line 244, it is written that the box model is constrained by the mean observed mixing ratio of BrO and IO, 2.5 and 1.4 pptv, respectively. What was the peak value of BrO and IO in the model runs?, if the peak value used was 2.5 and 1.4, did you run a sensitivity with the XO peak values measured by Read et al., 2008 and Mahajan et al., 2010 ?

The peak values used in the model simulations were 2.5 ppt BrO and 1.4 ppt IO. We have now performed a sensitivity analysis to these mixing ratios, using the upper and lower limits to the diurnal averages reported by Read et al. (3.5 ppt and 1.5 ppt for BrO and 2.0 ppt and 1.0 ppt for IO). The impacts of these changes on the diurnal profiles for OH and HO_2, and on the HO_2:OH ratio, are shown in the figure below (Figure S6). Given the focus of the manuscript on the different trends between the box and global models, rather than the absolute concentrations of BrO and IO, we propose to include this figure (Figure S6), which also includes sensitivity to early morning concentrations of BrO and IO as suggested by reviewer 1, in the supplementary information to the paper for completeness. We have added a comment in the main text (line 248) to refer to the sensitivity to BrO and IO concentrations ("*Sensitivity to these mixing ratios is discussed in the Supplementary Material*") and have added the following details to the Supplementary Material:

Observations of BrO and IO at the Cape Verde Atmospheric Observatory show average diurnal mixing ratios of 2.5 ppt and 1.4 ppt, respectively, which were used as constraints in the box model simulations presented in this work. In order to test the sensitivity of OH and HO_2 to these constraints, we have performed simulations in which the BrO and IO mixing ratios were constrained to the upper and lower limits of the observed values (3.5 ppt and 1.5 ppt for BrO and 2.0 ppt and 1.0 ppt for IO, as reported by Read et al. (2008) and Mahajan et al. (2010)). In addition, simulations were performed in which mixing ratios of BrO and IO increase from 0600 hours and reach the constant values of 2.5 ppt and 1.4 ppt, respectively, as shown by Read et al. (2008). Results from these simulations are shown in Figure S6, and indicate that there are only minor differences in the OH and HO_2 concentrations between simulations performed on constraining to the upper and lower limits of the observed BrO and IO concentrations, and that there is little sensitivity of OH or HO_2 to the early morning mixing ratios of BrO and IO.

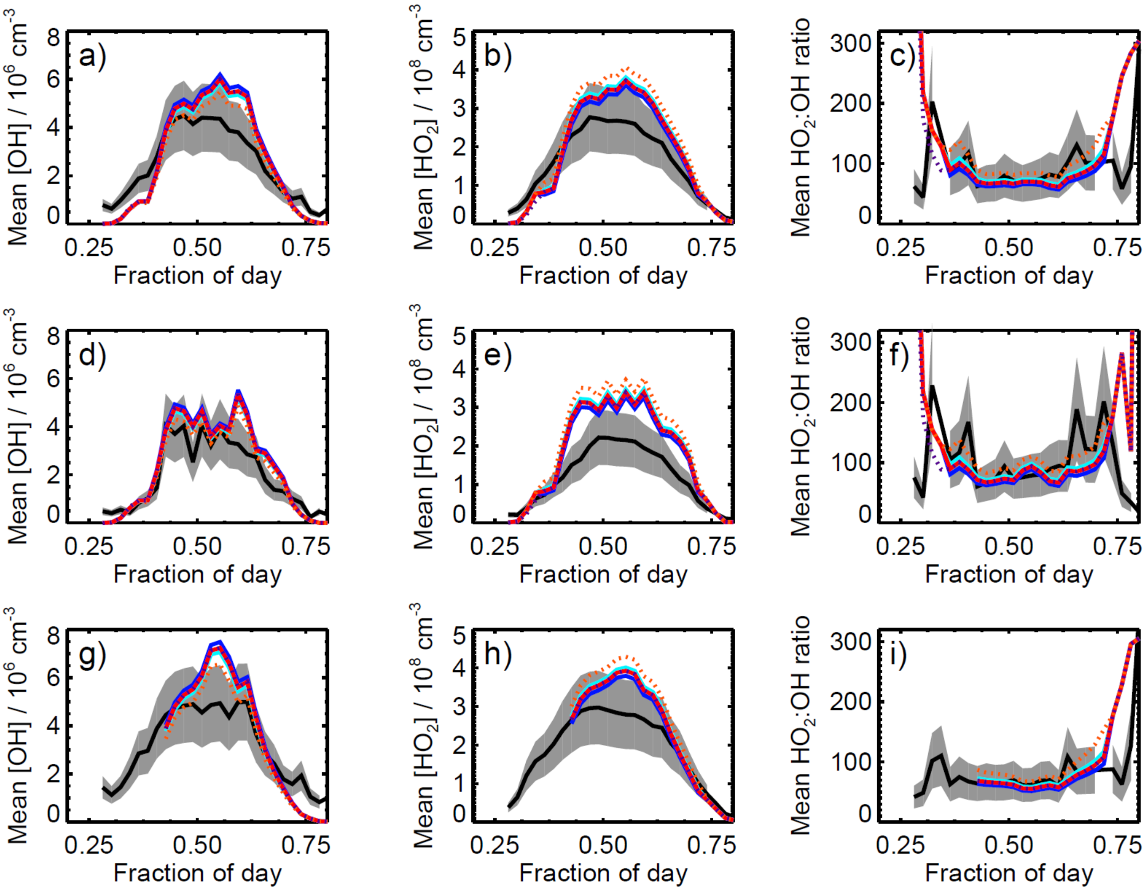

Figure S6: Impacts of changes to BrO and IO constraints on average diurnal profiles for a) OH during both measurement periods; b) HO_2 during both measurement periods; c) HO_2:OH ratio during both measurement periods; d) OH during SOS1 (Feb-Mar 2009); e) HO_2 during SOS1; f) HO_2:OH during SOS1; g) OH during SOS2 (May-June); h) HO_2 during SOS2; i) HO_2:OH ratio during SOS2. Observed data are shown in black, with grey shading indicating the variability in the observations; model simulations constrained to the average daytime (0930 to 1830 hours) mixing ratios of BrO (2.5 ppt) and IO (1.4 ppt) are shown in red; simulations constrained to the upper limits to the daytime mixing ratios of BrO (3.5 ppt) and IO (2.0 ppt) are shown in dark blue; simulations constrained to the lower limits to the daytime mixing ratios of BrO (1.5 ppt) and IO (1.0 ppt) are shown in light blue; simulations constrained to the average daytime mixing ratios of BrO (2.5 ppt) and IO (1.4 ppt) and including increases in the mixing ratios from 0600 hours are shown by the dashed purple lines; simulations with no halogens are shown by the dashed orange lines.

- One thing that I miss in this paper is the how halogen-driven changes in NO_x affect HO_x?. There is not mention to this aspect and from previous modelling studies it is expected to have a bearing on HO_x. I would suggest the authors to mention whether they have explored this coupling and perhaps add some additional results to the revised manuscript.

While we do not explicitly discuss the impacts of halogen-driven changes in NO_x on HO_x, this is included in the model simulations and there are some details in the discussion of the radical budgets and in Figures 4-6 which show how the role of NO_x chemistry in controlling radical budgets changes on inclusion of halogen chemistry. We propose to include the following details to discuss these impacts more thoroughly:

The change in the relative importance of HO_2 + NO on inclusion of halogens in the model results from both the increase in the total HO_2 sink, owing to the additional losses through HO_2 + BrO and HO_2 + IO, and the shift in NO_x partitioning owing to the reactions Br + NO → Br + NO_2 and IO + NO → I + NO_2. The reactions of

BrO and IO with NO result in a change in the NO_2:NO ratio of approximately 10 %, on average, which reduces the impact of HO_2 + NO as both a sink for HO_2 and a source for OH.

Minor Comments: P2,L54: " in in HO_x..", please remove one "in".

We have corrected this.

P3,L81: please replace "troposphere" by tropospheric?

We have changed this sentence to:

In general, observationally constrained box model simulations suggest that halogens in the troposphere will increase OH concentrations.

P31,Fig.1: For clarity, please improve quality in the box model results, e.g. what represents the yellow color?

The yellow data points represent the box model concentrations including halogen chemistry. We have corrected the figure caption, which previously stated the points to be orange, and have made some improvements to the presentation of the figure (symbol size, scale) to improve the clarity. Additional plots also are provided in the supplementary material to show these data in more detail