

Response to Anonymous Referee #2

With the aim of studying the sulfur- and halogen-rich emissions of Ambrym volcano (Vanuatu), the authors have included volcanic reactive halogen chemistry in a chemistry-transport model. This is an important effort and step forward in the study of volcanic reactive halogen chemistry, which has been restricted so far to 1-D chemistry models essentially. Ambrym volcano is a well-chosen case-study as it represents one of the most important source of persistent volcanic degassing on Earth, with substantial bromine emissions. This makes Ambrym an appropriate target for a first 3D modeling attempt. On the other hand, volcanic BrO observations are still sparse as BrO abundance is most often below satellite detection and consequently requires ground-based measurements to be evaluated. Regarding Ambrym volcano, ground-based observations of both sulfur dioxide (SO₂) and bromine oxide (BrO) column amounts, at various distances downwind from the crater are available making this volcano a good candidate.

This paper starts with an analysis of the BrO formation process, leading the authors to confirm previous findings achieved by 1D-models : they highlight the importance of model initialisation with high-temperature chemistry at the vent ; they also recognize that the highest SO₂/BrO values are located at the edge of the plume because of enhanced mixing with ozone-rich background air.

In the following part, the authors attempt a comparison of their model outputs against observations, with the aim of fitting the measured SO₂ and BrO abundances. Model outputs show a large underestimation of predicted BrO abundance by a factor of about 3.

The reviewer's comments helped us understand that the 'factor 3' discrepancy between model and DOAS observations for BrO columns was too much over-emphasized in our original manuscript. Indeed this factor refers to the ratio between the maximal DOAS BrO columns measured in the near downwind (~ 15 km from the vents) plume and its modeled counterpart. The model indeed underestimates by 60% in average the magnitude of observed BrO columns in the closest transect at 15 km from the vents. But, the model is in much better agreement with the DOAS observations made in the furthest transect (~40 km from the vents). In this case, the mean difference between observation and model is only 14% (relative to the mean observation). Overall, we find that the model underestimates the BrO DOAS observations by 40 % on average. This was already indicated in section 3.1.2. when Table 5 was discussed. We have now modified the Abstract, the main text (Sections 3.1.2, 3.2, 3.3.3) and Conclusions to more precisely characterize the discrepancy between observed and modeled BrO columns. In addition, we have made clear that this bias does not significantly impact our regional impact analysis as it has already decreased to 14 % at 40 km.

The authors perform various sensitivity tests to analyse the impact of a number of parameters on the reactive bromine cycle (vertical depth of the plume, formation of NO_x by high temperature chemistry, sulfate aerosol density). These sensitivity tests show that none of these parameters can explain the large discrepancy reached between model and observations.

The authors conclude that the ozone depletion modeling scheme implemented in their model is the likely source of this discrepancy. They notice that all of the ozone gets consumed in their model, thereby limiting the formation of BrO in the near-downwind plume in the model. Unfortunately, the present analysis does not bring clues toward a solution to this problem. Instead of performing an analysis that largely reproduces pre-

vious findings, the authors should attempt to discuss the potential role played by the various factors involved in the ozone depletion modeling scheme (ingredients, reactions, feedbacks, etc), so as to point more specifically the likely processes responsible for this modeling bias.

1) As noted by the reviewer, our study is the first 3D regional model study of volcano degassing taking into account volcanic plume chemistry and in particular reactive halogen chemistry. Therefore, we think that it is necessary to investigate whether our newly developed 3D model is able to reproduce some salient features of plume chemistry revealed previously by observations and explained by 1D model studies (i.e. previous findings) such as: BrO/SO₂ trends with distance from the crater and across the plume and the need for the high temperature initialization to reproduce the kinetic of BrO formation.

2) However, we also believe that our work is a progress compared to previous 1D plume studies :

(i) we make a “point-by-point” comparison of the modeled BrO to DOAS observations in transects made across the plume at different distances downwind i.e. each measured data point is compared with its temporally and spatially interpolated model counterpart (Figure 3, 4 and Table 5). This allows a fully quantitative comparison. Previous 1D model studies have only made a quantitative comparison to reported bulk downwind BrO/SO₂ usually as an average of observations with relatively large error bar (e.g. Bobrowski et al., 2007, von Glasow et al., 2010; Roberts et al., 2014; Bobrowski et al., 2015) and made only a qualitative comparison to the reported increase in BrO/SO₂ at the plume edge. Our approach combining 3D high resolution simulations and “point-by point” comparison to the DOAS data is now emphasized in the Abstract, the Introduction and the Conclusion. Note that to avoid some repetitions, we had to make some slight additional modifications to the abstract.

(ii) In addition, we have the potential to make the link between local scale observations close to the volcanic source and regional scale observations given by satellite data. We show that simulated SO₂ columns, when initialized with the Ambrym SO₂ source strength estimate derived from the DOAS observations by Bani et al. (2009) and (2012), agree within 25 % with the SO₂ columns detected by OMI. This is now highlighted in the section 4.1.

3) Concerning more specifically, the discrepancy on BrO, that is now better characterized in the manuscript as explained above:

(i) We do not agree with the sentence : *“The authors conclude that the ozone depletion modeling scheme implemented in their model is the likely source of this discrepancy”*

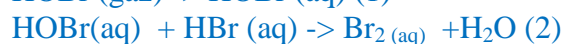
Indeed, the chemistry reaction scheme for BrO formation and ozone depletion in volcanic plumes is relatively well established (e.g., Roberts et al., 2009; von Glasow et al., 2009; Von Glasow, 2010; Roberts et al., 2014a.). Most of the gas and photolytic reactions are well known. Note that there are some uncertainties in Brx-NOx coupling. We choose not to include BrNO₂ following the findings of Roberts et al. (2014a) who includes a detailed BrNO₂ chemistry. Indeed, they find that BrNO₂ does not build up in the plume in contrast to von Glasow (2010) who had a more simplified BrNO₂ chemistry.

There are also some uncertainties in the gas-aerosol reactions as highlighted by Roberts et al. (2014b) but our sensitivity study varying aerosol loading (that can alternatively be seen as equivalent to keeping aerosol constant and varying the gas-aerosol reaction) showed no

improvement to the model-observation comparison. This was added in section 3.3.3 (see further below).

Instead, we conclude that the lack of ozone is possibly the reason for discrepancy as ozone is a requirement for BrO formation and is also destroyed during the BrO formation.

The following key set of reactions takes place in volcanic plume chemistry, as we now explain in the manuscript:



Because BrO can be photolyzed and the resulting O quickly react with O₂ to give O₃, the key ozone destruction steps are reaction 5 together with reactions 6, 7, 8, 9 (von Glasow et al., 2009). In strong (i.e. near-source and under high emissions) volcanic plumes, gas-phase cycling between Br and BrO (reactions, 5, 7 and 8) is a particularly important cause of ozone depletion. The subsequent lowering of ozone concentration limits the partitioning of BrO from Br. Hence BrO concentrations must become limited by lack of ozone. This is particularly important in the core of the plume, where there is less mixing with background air.

We have now added a section (section 2.1.2) where volcanic plume chemistry is briefly presented to help the reader follow our analysis of simulated plume chemistry and understand the limiting factors on BrO formation. We have also added a paragraph at the end of section 3.2 where we explain in detail the limitation on BrO due to the lack of ozone and refer to the reactions presented in the new section 2.1.2.

(ii) It is important to note that Bobrowski et al. (2015) also found in their model 1D runs of Nyiragongo's plume that BrO formation is ozone-limited in the concentrated plume. They also conclude that BrO and SO₂ do not provide enough information to test model simulations and they claim that measurement of ozone should be a priority for next measurements campaigns. We have added these comments at the end of section 3.3 (sensitivity studies).

(iii) We have now better explained our rationale in section 3.3 dedicated to sensitivity studies.

Our analysis based on model results, in particular Br speciation along the plume in the core and at the edges, indicates that BrO formation is limited due to ozone depletion. But, we have made some assumptions in the modeling of certain processes due to the lack of information to constrain the model or technical limitations. Therefore, we test whether the uncertainties in the representation of these processes could affect the model results and explain the discrepancies found between simulations and observations for the closest transects (~15-20 km) from the vents. More specifically, we focus on the depth of the plume that controls the degree of vertical mixing between emissions and background air, the formation of

NO_x by the high temperature chemistry and the surface aerosol area that can affect the in-plume chemistry.

In addition, we have rewritten most of section 3.3.1 (sensitivity to vertical depth) and 3.3.3 (sensitivity to aerosol loading) to clarify these studies. In particular, in section 3.3.1, we explain that increasing the vertical depth of the plume is equivalent to increase the degree of mixing between background air and volcanic emissions.

In section 3.3.3, we have also now mentioned that this sensitivity study is equivalent to increase the HOBr reactive uptake coefficient on sulfate aerosols, while keeping constant the aerosol surface area, for which large uncertainties exist. As mentioned by Reviewer 1, large uncertainties exist on heterogeneous chemistry. Note that the description of the simulated aerosol surface density has now been moved to Section 2.3.2.

(iv) We have also added a paragraph at the end of section 3.3 on an additional sensitivity study that we did not perform but that was done in two other studies and that can be interesting to our study. It is the role of total bromine emissions.

Here is this new paragraph

“Note that we did not test the sensitivity of the model results to the strength of total bromine emissions. Increasing total bromine emissions would increase total bromine in the plume. But because of the ozone limitation, this would lead to a reduced fraction of BrO and an increased fraction of Br. Finally, these two effects would compensate as found with a 1D model for the Nyiragongo’s plume by Bobrowski et al. (2015). As a result, increasing total bromine emissions would not impact BrO columns. This compensation was also found by Roberts et al. (2014) with their 1D model when compared their “high” and “medium” total bromine scenarios.”

(v) Concerning the comment of the reviewer : we could not find a solution to the discrepancy. We consider that it is a challenging problem to simulate volcanic BrO. Compared to previous studies, our model-observation comparison is particularly quantitative and in a way can highlight more the discrepancies. In addition, with a 3 D model, transport, mixing and concentrations in background air are not tuned by the user as they are in 1D models. We consider that we went as far as we could with these sensitivity studies. It is important to note that our study suffers from a lack measurements other than BrO and SO₂, even though our choice of Ambrym 2005 as a case study makes use of rather unique aircraft dataset of DOAS transects up to 40 km downwind. The main point is that BrO and SO₂ are not sufficient to fully constrain our modeling of volcanic plume chemistry as also highlighted in Bobrowski et al. (2015). This was already highlighted in the Conclusions but we have added it at the end of section 3.3.

(vi) We also consider it is important to mention that inaccuracies in DOAS retrievals can also affect the comparison. Under volcanic plume conditions (especially strong plumes as found during this extreme degassing event and close to the vents), it is well known that DOAS retrievals can suffer from inaccuracies. These have been estimated to some extent (e.g. Kern et al. 2010; Kern et al., 2012, Bobowski et al., 2010), focusing on SO₂. There remains a need to better understand DOAS uncertainties especially also for BrO.

In a second part (section 4), the impact of Ambrym sulfur- and halogen-rich emissions on sulfate aerosol, bromine and ozone content (section 4.2) at regional scale is proposed, as well as their impact on the lifetimes of methane and sulfate aerosols (section 4.3). However, it is difficult for the reader to evaluate the relevance of these regional analyses for multiple reasons :

- First, the uncertainty on the BrO content found in the first section, which is substantial given the three-fold discrepancy between modelled and observed BrO, is not put forward neither discussed to evaluate uncertainties on results at a regional scale.

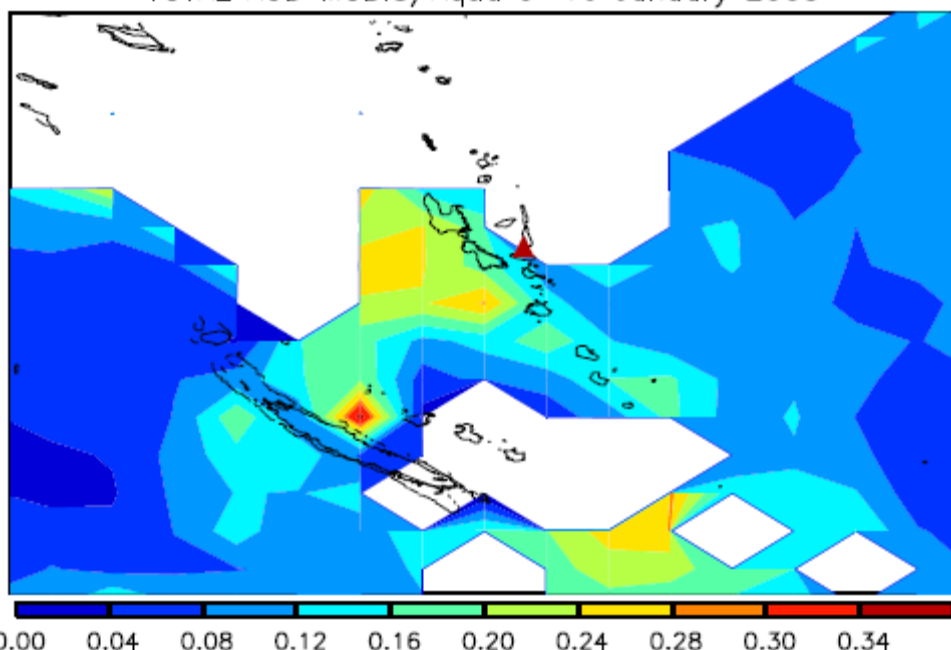
As mentioned earlier, we find a three-fold discrepancy when we compare the maximal BrO columns measured around 15 km of the vents with the corresponding modeled counterparts. Overall, the mean difference between BrO columns reported by Bani et al. (2009) and those simulated in our main simulation S1_HighT is about 40%, as indicated in section 3.1.2 and Table 5. For the furthest transect (40 km of the vents), the mean difference between observed and modeled columns is only of 14%. As a result, we believe that this small bias would not significantly impair the regional impact study. This is now explicitly stated at the end of Section 3 before studying the impact of the Ambrym volcano at the regional scale.

- Second, this section does not present any observation which could allow for testing the robustness (not to say the reliability) of these results. Satellite observations of aerosols (such as MODIS, POLDER, etc...) could be explored to better constrain model results.

- (i) We agree with the reviewer that the scarcity of data is a major challenge in the study of volcanic halogen Ambrym impacts. Nevertheless, in the section 4.1, we had presented a comparison between OMI SO₂ columns on 12th of January 2005 at 02:30 UT and the spatially and temporally interpolated model counterparts (Figure 10). We have now modified the text in the section 4.1 to present a more quantitative comparison. In particular, we have now calculated statistical quantities as the figure of merit in space (FMS) that quantifies the degree of spatial matching between observed and simulated plumes and the bias between the magnitudes of observed and modelled mean SO₂ columns. This comparison shows that we are able to reproduce fairly well the direction and the width of the plume at the regional scale as well as in good extent the magnitude of the SO₂ columns and give better confidence to our modeling.
- (ii) We agree that satellite data are available regarding sulfate aerosol. In the section 4.2, we have now added a plot showing the Total Aerosol Optical Depth from MODIS/aqua at 0.55 microns (see below). We have used the eight day average gridded L3 product from MODIS/Aqua (MYD08_E3) for 9-16 January 2005. We have added a paragraph in section 4.2 : “Total aerosol optical depth (AOD) at 550 nm is also shown in Figure 5S of supplementary material for the 9-16 January 2005. Enhanced AOD are clearly seen southeast of Ambrym in the direction taken by the plume the 12th January 2005 as discussed earlier (see Figure10) as well as northwest of Ambrym in the direction of trades winds. The latter point is again consistent with OMI SO₂ images from GSFC (Goddard Space Flight Center) at http://so2.gsfc.nasa.gov/pix/daily/0105/vanuatu_0105z.html showing that the plume was carried toward the northwest on the 14th and 15th January 2005. Enhanced AOD values varies between 0.12 and 0.34, which are approximately twice higher than the 3 years average (Oct.2005-Oct.2008) AOD presented by Lefevre et al. (2015). This is consistent with the extreme passive degassing activity of Ambrym during January 2005. This confirms the strong influence of Ambrym on the budget of

sulfate aerosol in the South West Pacific region and is qualitatively in agreement with our

TOTAL AOD MODIS/Aqua 9–16 January 2005



results. “

- Thirdly, according to the abstract and section names, the reader would expect that this second section would consist in an evaluation of the longer-term regional impact of Ambrym emissions that would generalize the study performed in the first part of the paper for a single day of emission (12 Jan 2005). However, Section 4 is only restricted to the same single event/day. Instead, the authors may broaden the scope of the study by assessing the impact of the continuous emissions of Ambrym.

- (i) We agree that a full assessment of regional scale impacts cannot be achieved by analysis of one day of simulation (there were 11 days of spin-up). But, we had strong computational limitations. Indeed, our simulations that include detailed chemistry and several nested grids are very demanding in term of computing time. We also believe that our study that includes model development, model evaluation at the plume level, sensitivity tests, a first investigation of plume chemistry on the regional scale and its implications for the troposphere was a substantial undertaking. For these reasons to make longer-term model runs of many months/years for a full assessment of impacts (to be repeatedly undertaken for a sensitivity study) would be a substantial second piece of work.
- (ii) Moreover, our paper confirms that the impact of Ambrym is regional and not only local in term of sulfur compounds. But it also stresses the role of halogen reactive chemistry at the regional scale. Indeed, we calculate significant bromine enhancement and ozone depletion at the regional scale up to thousand of kilometers of Ambrym. We have also shown evidence for bromine transport into the upper troposphere by convection. The influence of reactive halogen chemistry on the oxidizing power of the atmosphere is also demonstrated as well as on the lifetime of volcanic SO_2 and hence on sulfate production. This leads us to conclude that these halogen volcanic emissions and their associated chemistry need to be considered when studying the influence of volcanic emissions on climate. This is a point which is important and not yet well recognized in the atmospheric-volcanic community. Our study provides the evidence to motivate and guide future halogen-chemistry-climate assessments.

- Finally, the Vanuatu region is often cloudy. The formation of sulfate aerosol in

aqueous-phase may not be negligible in this context. However, this process is not included in the model. The authors should mention this potential issue, which may significantly impact the modeling results.

We agree that the formation of sulfate in aqueous phase might be important, and that this is a limitation of our study. This issue was already mentioned in the manuscript:

Section 2.3.1 “The sulfur scheme includes gas-phase oxidation, and dry and wet deposition, but not aqueous-phase oxidation.”

Section 4.2.1 “In this model study, the aqueous-phase oxidation of SO₂ to sulfate was not taken into account. This process becomes self-limiting in strong volcanic plumes due to the titration of oxidants for example H₂O₂ (Schmidt et al., 2010) but may have a significant contribution to sulfate formation over the whole model domain thus will be considered in future work.”

Section 4.4 “Our regional 3D model study includes a less detailed SO₂-sulfate chemistry scheme (gas-phase oxidation only) but includes detailed plume reactive halogen chemistry.”

This is further emphasized in the revised version of Section 4 and Conclusions as a model limitation and an area of future improvement. Specifically we have now added, as the reviewer mentioned, that this process may be of particular importance because the Vanuatu region is particularly cloudy (provided the aqueous-oxidation is not self-limiting i.e. in relatively dilute plumes)

While significant effort has been undertaken by the authors to include reactive halogen chemistry in a 3D chemistry-transport model, the manuscript is lengthy and relatively difficult to follow for the reader. According to me, this article would benefit to be divided in two papers (possibly a companion paper).

- The first paper would require more developments on the modeling aspects in order to find a better first-order agreement between modeled and observed downwind BrO abundances, which represents the critical observations of this study. At least should the model processes responsible for model biases be listed and discussed in details.

We have already replied above to all these aspects. We summarize here our answer.

First, overall the mean difference between BrO columns reported by Bani et al. (2009) and those simulated in S1_HighT is about 40% (relative to the observations) as indicated in section 3.1.2. We consider that this is a reasonable result under the conditions of our study.

The bias is more pronounced (60%) for the transect closest to the source but small further downwind around 14%.

Secondly, with the model, we have done a thorough analysis of the plume chemistry by looking at Br speciation evolution in the plume at the edge and in the core that leads us to identify that the lack of ozone in the simulation in the plume limits the partitioning of BrO from Br as explained above.

Thirdly, we have tested whether a misrepresentation of some model processes (due to a lack of information) could be the reason for the discrepancy. We had identified several processes whose modeling is uncertain and could impact BrO. It was injection height, model mixing, NO_x emissions due to high temperature chemistry and aerosol loading. For this latter process, the sensitivity study was equivalent to perform a sensitivity study on the value of the uptake coefficient. We have also added a discussion on the impact of total bromine emissions.

So, we have ruled out the implications of these different processes to explain the discrepancy. We consider that we went as far as we could. We also conclude that measurements of BrO and SO₂ are not sufficient to fully constrain our modeling of volcanic plume chemistry.

The improvements made in the paper in response to the reviewer were described above when we answered in detail to each question.

- The second paper would require more constraining observations to validate results of the impact of halogen-rich emissions at a regional scale. As Ambrym is continuously degassing, a longer-term study would be possible, i.e. not restricted to a single day of substantial emissions. This would provide a global and more representative estimation of the actual regional impact of Ambrym emissions.

We have also replied to this comment above. We summarize our answer here. We agree with the reviewer that a long-term study would be necessary to fully understand the actual regional impact of Ambrym. But it would be a substantial second piece of work, given that our simulations are very demanding in term of computing time. We also think that our study (model development, model evaluation at the plume level, sensitivity tests, a first investigation of plume chemistry on the regional scale and its implications for the troposphere) is already a substantial undertaking. Our study also highlights interesting results on the regional influence of Ambrym. In particular, it emphasizes that reactive halogen chemistry should be considered when studying the impact of volcanic emissions on climate. We have given more details above in our previous answer.

Minor comments :

- Page 35324: sentence in line 1 has to be rephrased.

The sentence was replaced by “Biogenic emissions were provided by a monthly mean climatology for the year 2000 produced with the MEGAN (Model of Emissions of Gases and Aerosols from Nature) database (Guenther et al., 2006).”

- Page 35326, line 23 : ‘Due to

...

’ : sentence not ended

It was corrected. The sentence is now : “Due to uncertainty in volcanic NO_x emissions (see discussions of Martin et al., 2012; Roberts et al., 2014a; Surl et al., 2015), HSC Chemistry output both with and without NO_x were used to initialise CCATT-BRAMS (Simulations S1_HighT and S1_HighT_noNO_x).”

- Page 35332, line 5 : ‘similar to’ (not ‘than’)

We believe that that the sentence was on page 35331 line5. We have made the correction.

- Page 35334, line 26 : remove first ‘reactive’

This was removed.

- Page 35336, lines 26-28 : which are the radicals other than NO_x that you think are important ?

We have tested the impact of NO_x emissions on the plume chemistry by comparing two simulations. The first one S1_HighT includes emissions of OH, NO, Cl, Br to take into account high temperature chemistry at the vent in addition to emissions of SO₂, sulfate aerosols and halides HCl and HBr. The second S1_HighT_noNO_x has the same emissions than S1_HighT except that it does not include NO emissions. So by “other radicals” we meant OH, Cl and Br radicals.

This is now clarified earlier in the paragraph. The sentence (previously p35336 1 18-20) “We performed a simulation (S1_HighT_noNO_x) where from the high-temperature initialization was not included while keeping constant the emissions of the other radicals from HSC” was replaced by this sentence “We performed a simulation (S1_HighT_noNO_x) where NO_x from the high-temperature initialization was not included while keeping constant the emissions of the other radicals (i.e. OH, Cl, Br) from HSC Chemistry.”

The sentence (previously p35336, line 26-28) mentioned by the reviewer became :
“This suggests that NO_x emissions are not crucial to kick off the chemistry initially but that they are responsible for the decline of HBr further downwind after 15 km.”

- Page 35340 : *Could you explain more why the result on sulfate aerosol burden confirms that sulfate which formed from atmospheric oxidation are the main driver of volcanic halogen chemistry ?*

We find that the sulfate burden has increased by 0.08 Tg due to Ambrym emissions since the beginning of the simulation on January 12th. This was calculated by comparing the sulfate burden between S1_HigT and S0. But the total direct emission of sulfate due to Ambrym since the beginning of the simulation reaches 3.34 Gg on January 12th. This cumulated direct emission represents 4% (= 3.34Gg/0.080Tg) of the total burden. It is a maximal value, as these emissions could have been washed out in the model or left the domain of the study.

To clarify, we have modified the sentence “This means that at least 96 % of the sulfate burden increase due to Ambrym results from the atmospheric oxidation of SO₂ from the volcano by OH.” by “This means that at least 96 % \approx (80 -3.34)/80x100) of the sulfate burden increase due to Ambrym results from the atmospheric oxidation of SO₂ from the volcano by OH.

- Page 35340, line 14 : *replace ‘sulfate is’ by ‘sulfate aerosols are’*

This was corrected.

- Page 35340, line 22 : *words are attached here ‘HighTand’ but also in several places throughout the text.*

This was corrected.

- Table 4 : *what are the sources used to determine the ratios used to initialise the model?*

Ratios from Table 4 are explained in detail in Section 2.3.3:

- HBr/SO₂ and HCl/SO₂ ratios are derived from measurements of Allard et al. (2009).
- H₂SO₄/SO₂ ratios are derived assuming that 1% of sulfur is emitted as H₂SO₄ based on Mather et al. (2003) and on Von Glasow et al. (2009).
- OH/SO₂, NO/SO₂, Cl/SO₂, Br/SO₂ are output from an HSC chemistry simulation initialised with data from Table 3 as explained in Section 2.3.3 where references are given.

We think that we cannot put all these information in the caption of the Table 4. But, we have added “See section 2.3.3 for the detailed explication of the ratios derivation” in the caption of the Table 4. For clarity, we have also rephrased some of the sentences of the Table 4.

We have also added in Table3 that mixing ratios are mass mixing ratios.

Additional corrections from the authors in Abstract, section 4.3 and Conclusion :

We have rephrased the results of section 4.3. Because of the non linearity of the chemistry, we cannot say: “Reactive halogen chemistry is responsible for about 62% of the methane lifetime increase with respect to OH, with depletion of OH by SO₂ oxidation responsible for the remainder (38%).” Indeed, it is not strictly possible to determine the contribution of one process (for example reactive halogen chemistry) by turning it off and compare with a simulation where the process is taken into account. Indeed, the contribution that we find by doing this, for instance 62% here, is not necessarily right because of the non linearity of the chemical system. So we replace the sentence in the abstract the sentence by “When considering reactive halogen chemistry, the lengthening of methane lifetime with respect to OH is increased by a factor of 2.6 in our simulation compared to a simulation including only

SO₂ emissions” . Similar sentences are now in the main text (section 4.3), Abstract and Conclusion.

For the same reason, we have rephrased “The reactive halogen chemistry in the plume is also responsible for an increase of 36% of the SO₂ lifetime with respect to oxidation by OH” by “Including the reactive halogen chemistry in our simulation increases the lifetime of SO₂ in the plume with respect to oxidation by OH by 36% compared to a simulation including only volcanic SO₂ emissions”.