Response to Anonymous Referee #1

This paper presents a thorough multi-scale chemistry-transport modeling analysis of a volcanic plume from the Ambrym volcano (Vanuatu). Model simulations are evaluated against ground-based and satellite observations of SO2 and BrO. A lot of care is put in estimating as accurately as possible model inputs, notably the mix (sulphur, halogen), vertical distribution and rates of volcanic emissions. The authors set up a complex model configuration to generate a realistic transport of volcanic emissions. In order to resolve the plume at different scales and phases, the atmospheric model domain is 3 nested grids centred over the volcano going to a resolution of 500 x 500 m; ECMWF meteorological analyses (wind, temperature, water vapour) are used to initialize and nudge the model. A range of model simulations is performed to cover the uncertainties in model inputs and chemistry (e.g. high temperature chemistry, plume height,:::
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The introduction on volcanic halogen is rather thorough. However, it does mention the issue of the transport of volcanic halogen to the stratosphere and the possible implications for stratospheric ozone. It is a bit surprising because this point pops out a few times in the manuscript. For example, it is mentioned in section 4.2.2: "Such transport of volcanic bromine to the upper troposphere and the stratosphere is of strong interest. Indeed, the stratospheric bromine burden is underestimated by global models that take only into account long lived halons and methyl bromide". Again, in the conclusion, the authors recall an important finding in their model simulations: "There is also evidence in the simulations of a subsequent transport of bromine to the stratosphere from Ambrym". They also state that "longer duration simulations should be performed to fully quantify the impact of Ambrym on chemical composition of the troposphere at the regional scale. In particular, flux of bromine to upper troposphere and to the stratosphere from this extreme continuous degassing event". They never provide references and this point is not mentioned in the introduction. The references are about the impact of volcanic halogen on tropospheric chemistry. I would suggest to mention the impact of volcanic halogens on the stratosphere, notably the ozone layer, in the introduction with references. This issue has been largely overlooked in the literature. But, several recent studies have shown that volcanic halogen, notably bromine, from very large volcanic events could have had a drastic effect on the evolution of stratospheric ozone in the past, when the atmospheric chlorine and bromine loadings were low (in contrast to the present-day atmosphere). The key uncertainty in assessing the impact of volcanic halogen from massive eruptions on stratospheric ozone is the fraction of bromine and chlorine emissions reaching the stratosphere which depend on the form of the volcanic halogen injected. If bromine and chlorine are in the form of HBr and HCl (acidic molecules, soluble), they would tend to be eliminated very quickly within the plume by dissolution in aqueous phases ou/and adsorption on solid particles. In contrast, bromine and chlorine radicals are much less soluble, so they would survive longer in the volcanic plume and hence are more likely to reach the stratosphere. Therefore, the process of halogen activation in volcanic plumes is highly relevant to the question of the potential impact of volcanic halogen on stratospheric ozone.

We agree with the reviewer's comment. We have now added the impact of volcanic halogens emitted by explosive eruptions on stratospheric ozone (with the references to relevant articles) in a paragraph in the Introduction :

"On the other hand, the impact of other compounds as halides (HCl, HBr) injected by explosive eruptions into the stratosphere as well as the overall impact of minor eruptions and quiescent passive degassing have been largely overlooked. However, the presence of volcanic halogens in the stratosphere following explosive eruptions has been recently detected (e.g., Hunton et al., 2005; Rose et al., 2006; Prata et al., 2007; Theys et al., 2014, Carn et al., 2016). Such volcanic halogen injection, enabled by incomplete volcanic halides washout as first predicted by a model study (Textor et al., 2003), was found to cause enhanced reactive chlorine and bromine, Polar Stratospheric Cloud formation as well as enhanced ozone depletion (Rose et al., 2006; Millard et al., 2006). As a result, it is important, as emphasized in Cadoux et al. (2015), to consider volcanic halogens in addition to sulfur compounds when studying the influence on the stratosphere of past and future explosive eruptions."

We have also mentioned in the Conclusion the potential impact of halogen activation in tropospheric plumes on stratospheric ozone :
"There is also evidence in the simulations of a subsequent transport of bromine to the stratosphere from Ambrym. Thus, the halogen activation in tropospheric volcanic plumes could be one aspect of the potential impact of volcanic halogen on stratospheric ozone."

The first part of the paper is devoted to the analysis of near field plume focusing on the higher resolution model nests. Overall, the results are encouraging. Comparisons between simulations and ground-based/satellite SO2 observations indicate that the model performs rather well regarding the transport and dispersion of the plume. Sensitivity simulations confirms that the high temperature chemistry in the vent of the volcano is important because, by taking it into account, the model is able to reproduce the general evolution of BrO/SO2 seen in DOAD data downwind from the vent. The main discrepancy is about the magnitude of BrO columns that the model underestimates by about a factor 3. The authors point out that the fact that BrO formation is limited by the flux of ozone in the near-downwind plume could explain some of the discrepancy. Unlike a 1-D plume model, the level of mixing between the plume and the ambient air is a factor whose influence they cannot easily explore in their 3-D model configuration. Plume chemistry and the associated changes in chemical composition strongly depend on the entrainment flux of outside air. Clearly, the different changes, notably ozone destruction and BrO formation, do not have the same dependency on the mixing rate. For instance, the lower the mixing is, the more pronounced the local ozone destruction, but the lower the formation and hence the levels of BrO are. This discrepancy may indicate that the level of mixing is not quite right yet. The balance between mixing and chemistry is difficult to strike.
We agree with the reviewer's summary. In the section 3.3.1, we have modified the text to make it clear that through a sensitivity study on plume depth, we test the effect of plume-air mixing. We agree though that efforts to test plume-air mixing are more challenging with a full 3D model than with 1D models where the mixing is to a large extent user-tuned.

Anyway, I think the authors went as far as possible with their model configuration. Another possible cause for the discrepancy is the halogen activation scheme because there are large uncertainties pertaining to aqueous phase chemistry.
This point is now mentioned in the manuscript in the section 3.3.3. In this section, we have performed a sensitivity study where we have increased the sulfate aerosol surface density. This is equivalent to keep the aerosol surface density constant but to increase the reactive uptake coefficients of HOBr and $BrONO_2$. Indeed, there are a lot of uncertainties on the values of uptake coefficients as shown in Roberts et al. (2014b) where HOBr +HBr and HOBr +HCl reactive uptake coefficient have been reevaluated.

It is also important to note, as mentioned elsewhere in our responses to Reviewer 2, that the discrepancy between observed and simulated BrO columns is now better characterized in the text. We find that 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%. Overall, we find that the model underestimates the BrO DOAS observations by 40 % in average. We have now made clear as well that this bias does not significantly impact our regional impact analysis as it has already decreased to 14 % at 40 km.

The second part of the paper is devoted to the analysis of far field plume focusing on the large-scale model domain where the resolution is 50x50 km. Large-scale model simulations are evaluated against SO2 columns. It is a pity that simultaneous BrO observations are not available. The authors discuss the model-calculated impact on the lifetimes and budget of several trace gases (e.g. ozone, methane, OH, SO2). All the results confirm that volcanic halogen emissions may play a significant role in atmospheric chemistry, at least on a regional scale.
In conclusion, I congratulate the authors for this solid piece of multi-scale volcanic plume modelling. This work is a big step forward from the simple 1-D plume approach. It allows a more critical testing of model simulations against observations because the transport and dispersion of the plume are much better constrained.
The model results confirm the potential importance of volcanic halogen on the budget of a range of trace gases. The magma of some volcano is extremely rich in halogen. Therefore, when assessing their impact on atmospheric composition and climate, the halogen component should be not neglected. I recommend publication. However, it might be good for the authors to take on board some of my comments and suggestions listed above.