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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modeling the reactive halogen plume from Ambrym volcano and its impact on the troposphere with the CCATT-BRAMS mesoscale model" *by* L. Jourdain et al.

Anonymous Referee #2

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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 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 NOx 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 previous 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.

In a second part (section 4), the impact of Ambrym sulfur- and halogen-rich emissions

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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.

- 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.

- 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.

- 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.

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

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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.

- 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.

Minor comments :

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

- Page 35326, line 23 : 'Due to. . . ' : sentence not ended

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

- Page 35334, line 26 : remove first 'reactive'

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

- 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 ?

- Page 35340, line 14 : replace 'sulfate is' by 'sulfate aerosols are'

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

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

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 35313, 2015.