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Interactive comment on "Towards a representation of halogen chemistry within volcanic plumes in a chemistry transport model" *by* L. Grellier et al.

Anonymous Referee #1

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1 Overview

This paper presents a modelling study of the 10 May 2008 eruption of Mount Etna. The authors use a series of box-models to investigate the evolution of the halogen chemistry within the volcanic plume over the course of the eruption and the following 18 hours. The sensitivity of this evolution to different parameters is investigated, and two different implementations of a "plume box" for processing of the very early plume are evaluated.

2 General comments

The model presented is a development towards a 3D Eulerian chemistry and transport model. The scientific questions regarding plume evolution have, in the literature, typically been addressed with Lagrangian models. However, a Lagrangian approach cannot be easily implemented into 3D models, so there is a need to develop methods for incorporating volcanic plumes within Eulerian models. The most significant challenge regarding this is the problem of incorporating a strong point source in a low resolution model –in such models the emissions are instantly diluted over the area of the grid used, which may prevent adequate modelling of non-linear processes within the plume.

Due to its very simple physics the model has significant flaws as an investigative tool, both in comparison to Lagrangian models and 3D Eularian chemistry and transport models. These flaws are discussed in the section below, and includes two serious issues that must be addressed before any publication. Despite reasonable agreement with the satellite measurement of BrO, these flaws are significant enough that it cannot be said that the model is a reliable simulation of this particular plume. At a push, the results may be considered a more generic approximate simulation of a plume-like environment. The investigative domain of the results, and the sensitivity studies conducted, explore new territory for volcanic plume modelling. In particular this is the first study to investigate the halogen chemistry of a volcanic plume during the night.

Does the manuscript represent a substantial contribution to modelling science within the scope of this journal? - Previous studies have investigated plumes using Lagrangian models. This model, while described in the text as a "1D-framework", is best described as a stacked box model. Due to the lack of transport (or any parametrisation thereof) this is inherently inferior to the Lagrangian models used previously in the literature. I can identify two main contributions to modelling science methods in this paper:

- The method of calculating the Br2/BrCl branching ratios (a minor, but interesting result)
- The "Plume 1" method of considering the very early plume. This method would be of interest to modellers considering how volcanic emissions could be implemented in a Eulerian model. (As discussed in the next section the "Plume 2" case is useless).

The paper fails to adequately present a conclusion regarding the second point above. For the case study, the method *appears* redundant as Br totally partitions to BrCl in the night. It is unclear if the authors intend for this publication to present a null result for the utility of the parametrisation scheme, however this is the conclusion that is likely to be drawn by the reader.

Are the scientific approach and applied methods valid? Do the models, technical advances and/or experiments described have the potential to perform calculations leading to significant scientific results? - While the model created appears to be a valid box model, the scientific question it is tasked to answer requires a consideration of transport and mixing beyond the capabilities of a box model.

To what extent is the modelling science reproducible? Is the description sufficiently complete and precise to allow reproduction of the science by fellow scientists (traceability of results)? - Yes, the description is adequate in this regard.

Are the scientific results and conclusions presented in a clear, concise, and well structured way (number and quality of figures/tables, appropriate use of English language)? - The paper is very long, and is, in general, overly-verbose in its descriptions. The Conclusions section contains a long description of the methods used and the planned future work.

The manuscript suffers from the central flaw that the model is inadequate to produce reliable results and is fundamentally inferior to prior Lagrangian models as a plume

chemistry investigation tool. The absence of transport makes this stacked box model simulation neither an reliable depiction of real plume chemistry nor the likely modelled chemistry that would occur within the volcano-containing box of a 3D model. This reviewer's recommendation would be to not publish this manuscript, but to wait until the 3D-MOCAGE is developed and to use the results in this manuscript to provide direction to that investigation.

3 Specific comments

There are two major specific problems with this paper:

1. The first is illustrated by Figure 4. It is shown that there is significant plume density at low levels, and the increase in BrO mixing ratio in the lowest levels suggests that this plume density is in the planetary boundary layer. This is non-physical. Mount Etna is roughly 3 km in altitude (700 hPa) and emits into the free troposphere. There is evidently an error in the assumed topology here with the volcano emitting from too low an altitude.
2. The second concerns the "Plume 2" runs. In these runs the volcanic emission is initially contained within a 0.025°$\times$ 0.025°(2 km $\times$ 2 km) plume box, which is not mixed with the the rest of the model box until the end of the 4 hour eruption period. This assumes that the plume would not disperse significantly more than this area over the course of the eruption, which is a very non-physical assumption. Gas emitted at the start of the eruption would likely have been advected a considerable distance (of order 100 km) over 4 hours by free tropospheric winds. The "Plume 2" scenario results in a very unrealistic, highly concentrated plume which does not have physical relevance. This scenario has no demonstrable relevance.

While not as major as point 2 above, there is a related issue regarding the general model box. The model grid size of 0.5°× 0.5°(approx. 45 km × 45 km) has a dimension smaller than the likely advection distance of the plume over the eruptive period. This may actually be counteracting another implicit inaccuracy; the dispersion in the direction perpendicular to plume travel is likely to be much less than 45 km. While the two errors may compensate and result in the model box being a reasonable estimate for the plume area, this issue should be addressed in the text.

Over the course of the eruption the emitted gases accumulate within the model grid box and the concentration of halogens increases over time. While this is an accurate reflection of what the average concentration over an area containing the erupting plume would do, it does not reflect the trend of halogen concentration in any part of the plume. In a Lagrangian framework, the core of the plume would have a maximum concentration of volcanic volatiles at the the point of emission and would fall over time as the plume dispersed. In general, the influence of the plume within any air parcel downwind of the volcano would be determined by mixing, rather than by the continuing eruption. Therefore the evolution of the plume chemistry discussed in section 5 of this paper cannot be considered a reasonable assumption of the evolution of any part of the plume. This should be acknowledged in the text where the chemical evolution of the plume during the first four hours is discussed and any identifications of chemical phenomena occurring in the eruptive period should be qualified with this caveat.

It is unfortunate that the end of the eruption (18:15) and the onset of night (18:30) are almost simultaneous. The authors direct the readers attention to changes in the chemistry that occur due to dynamic reasons, however for the reader it is difficult to distinguish these effects from those due to the approach of night. While this is justified as being the physical reality of the eruption, it would be highly informative to evaluate an eruption where there were several hours of daylight following the end of the eruption.

The collection of BrO and Br2 as a single BrOx species seems to introduce a potential problem. While Br2 would be a stable species in the night, the BrO + NO2 -> BrONO2

reaction can still occur at night time, effectively destroying this species. This is probably a minor concern in this study, as BrOx and HBr are almost totally destroyed before sunset anyway, however it could be a major issue in other circumstances. If this is addressed in the mechanism, this should be acknowledged in the text.

Reaction R8 generates Cl radicals. It is not explicitly stated what happens to these radicals, and as a large volume of BrCl is photolysed at dawn this could be important. If it is the same as Roberts (2009) this should be referenced.

4 Technical comments

- Page 2588, line 21: “Supplement” should not be capitalised. “and” should be changed to “*et*”
- Page 2588, line 25: “The eruptive cloud was injected up to 5 km in height". State whether this is 5 km above sea level or above the vent.
- Page 2600, line 25: Table 1 would be the gaseous composition leaving the *vent*, whilst the composition leaving the *crater* would be the figures in Table 2 (plus some in-crater dilution). Needs clarification here.
- Page 2601, line 10: hyphen needed between “one” and “dimensional”.
- Page 2603, line 7: “Volume” should be changed to “area”.
- Page 2604, line 9: “Argon” should be changed to Ar for consistency.
- Page 2606, line 2: “Table” should not be capitalised.
- Page 2606, line 14: “emission injected over 0 and 5 km” should be changed to “emission injected between 0 and 5 km above the crater”.

- Page 2606, line 22: “and also for sulphur dioxide” should be changed to “this is also the case for sulphur dioxide”.
- Page 2609, line 9: add “in” between "decrease” and “HBr”.
- Page 2609, line 15: change “less” to “fewer”
- Page 2610, line 15: remove “that”
- Page 2610, line 17: Add “on” between “05:00 UTC” and “the”
- Page 2615, line 20: Change “ration” to “ratio”
- Page 2617, line 11: Change “simulation duration” to “computational cost”
- Page 2618, line 3–7: Rather than multiplying by 16 to get an artificial figure, comment in the text that the 2°× 2°figure is a factor smaller than the 0.5°× 0.5°that is approximately equal to the ratio of the two areas involved.
- Page 2618, line 27: Saying the depletion of O3 is smaller in the 2°× 2°run is misleading. Although the fall in concentration is smaller, the total amount of O3 destroyed is similar-greater because this box is 16 times larger. Need to reword here.
- Page 2620, line 9: “oxidants” should be changed to “oxidant”
- Page 2620, line 24: “a field” should be changed to “afield”
- Table 2: “Espèces” should be changed to “species”
- Figure 4, caption: add “on” before “the 11 May”.
- Figures 5, 6, 8, 9 and 12 caption: text “of the emission levels” doesn’t make sense and should be removed.