Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-1110-RC2, 2020

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Co-emission of volcanic sulfur and halogens amplifies volcanic effective radiative forcing" *by* John Staunton-Sykes et al.

Anonymous Referee #2

Received and published: 16 December 2020

This study considers hypothetical VEI 6 and VEI 7 sized eruptions and, using a coupled chemistry-aerosol model driven by scenarios including or not including halogen injection, investigate how the co-emission of volcanic sulfur and halogens alters the evolution of the volcanic aerosol plume, stratospheric ozone chemistry, and the resulting radiative forcing and UV flux.

The authors investigate how volcanic halogens may interact with the sulfur aerosol life cycle and interact to modulate volcanic forcing conversely to previously reported work. I found the link between chemical and microphysical processes of particular interest. The authors reveal in their model experiments the primary importance of halogens in major volcanic emissions in the sulfur cycle in the stratosphere, a process already suspected for eruptions of much more minor amplitude. Impacts of halogen emissions

on dynamics of the aerosols and the subsequent effect on aerosol microphysics are also considered here when critically missing in previous reported studies. Effects on some key stratospheric compounds like ozone, water vapour and methane are also analysed. To me, this work points at the critical need to maintain space-borne observations of stratospheric compounds which will be particularly valuable to quantify halogen injected by volcanoes and needed for model initialization. The authors finally mention open questions to be addressed in future studies reflecting the great interest to consider these events and their associated various injections (sulfur, halogens, ash, water vapour) in the future climate.

This study is original and comprehensive judging by the various topics and impacts covered (microphysics, dynamics, chemistry and radiative forcing). I found the manuscript clear, well-written and nicely going straight to the obtained results. I estimate that this work deserves to be published in ACP after the following minor comments have been addressed.

Specific comments:

Introduction: I am not a specialist of petrological processes but could you indicate the degree of uncertainty when petrological budgets are used to derive stratospheric inputs of halogens? What are the assumptions behind the halogen injection efficiency (I would suggest to briefly recall the definition of the stratospheric halogen injection efficiency).

P5 lines 158-168: How the overall chemical species initialized? Is it based on climatological 3D fields or only surface emissions provided by CMIP6? Especially for bromine compounds, how the Bry budget initialized? Are very-short-lived species accounted for? The resulting inorganic bromine budget in the stratosphere and more generally the inorganic halogen content, computed in chemistry models is of particular importance regarding ozone chemical cycles and would have significant impact in your scenario with no volcanic emissions of halogens. Please provide a bit more information.

P5 line 170 onwards: You do not differentiate between SW and LW radiation conversely to the work of Schmidt et al. (2018). What justifies this choice?

P5 line 188: why variations in the surface albedo were not taken into account? As you state in the manuscript, the model can be forced by surface boundary conditions. Is it for calculation-time issues?

P6 line 202: An injection altitude distributed around 21 km has been chosen. Volcanic impact depends on injection altitude especially because the residence time of aerosols is linked to this parameter. I am aware that strict choices must be done for costly long-term simulations but what justifies this value? Did the authors conduct sensitivity tests on this parameter?

P6 lines 206-219: there is a lot of assumption behind the stratospheric halogen injection efficiency. The values given by Textor et al. (2013) strongly differ from other reported studies. Is this factor highly variable from one eruption to another? Why El Chichon and Mazama eruptions reassures the numbers taken for HAL56? For HAL10 it is not clear to me why the Pinatubo HCL:SO2 molar ration must match the one for Mt Mazama. I guess petrological processes somewhat differ for these events. Please clarify.

P8 figure 1d: what is the reason for the overlapping HAL and SULF S global burden at an early stage of the simulations? This surprises me because this feature is not visible on SO4 which already shows a marked difference over the first months (figure 1c).

P11 lines 268-279: The investigation of Reff is interesting since it provides a (integrated) description of the impact of HAL scenario on aerosol sizes. However, it would have been also valuable to examine more comprehensively the impact on microphysics. Although GLOMAP is a modal microphysical module (as far as I understood) did the authors get information about the effects on size distributions (geometrical standard deviation, total concentration)? For instance, concentrations might be reduced if particle sizes increase but with different ratios. Concentration (well, the whole size dis-

tribution) is also important for the SAOD and ERF calculated in the manuscript (figure 4).

P11 lines 268-279: I think a short comparison with maximum Reff values reported for different past eruptions (Pinatubo in particular) would be interesting to include here.

P11 line 291: please add the wavelength here.

P12 figure 4: the color coding (red for SULF and blue for HAL) is the opposite than in previous figures. I think it would be preferable to homogenise this.

P14 figure 6: A latitudinal transport is visible in the lower stratosphere, particularly for HAL10 simulation. What process can be related to the hemispherical difference? Since the plot is integrated over 3-years I am not sure that the dominating phase of the QBO (which has been shown to impact volcanic aerosol transport from the tropics) can be an explanation.

P15 lines 360-366: I guess the methane increase, although limited, for the sulfur-only scenario is chemically due to the reaction of CH4 + OH. This would mean that less OH is present under volcanically-impacted periods. There is a complex interplay between HOx, nitrogen and halogen chemistry that can result in OH reduction (and CH4 increase) unless the dominating process deals with the very high amounts of SO2 that may sequester OH through reaction SO2+OH (subsequently leading to the formation of sulfuric acid). In my mind, OH was rather increased for summertime midlatitude eruptions (as shown for the 2009 Sarychev eruption) reflecting a possible seasonal effect. Changes in methane amounts are also likely resulting from radiative/dynamical origin with more troposphere-to-stratosphere transport resulting from the aerosol heating in the tropopause region. A significant part of ozone changes following major eruption has been attributed to changes in transport (see e.g. Pitari, G. and Rizi, V.: An estimate of the chemical and radiative perturbation of stratospheric ozone following the eruption of Mt. Pinatubo, J. Atmos. Sci., 50, 3260–3276, 1993). Similar process could apply for methane. Do the authors have an idea about the process behind the CH4

slight increase?

P18 line 404: I found the demonstration about ozone change (although largely trusted) as the dominant driver of ERFclear, clean a bit abrupt. Please provide more details here about the method used rather than only citing the Rap et al. reference.

P20 line 461: specify “for two winters”.

P21 figure 12: this figure (as figure S4) is very interesting but I think plotting anomalies (by subtracting each simulation with the control run) would have been more meaningful especially to highlight the effect of SULF and HAL scenarios on the NH high latitudes. Such figure could be added in the supplementary material.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-1110, 2020.