RC1: 'Comment on egusphere-2023-184', Anonymous Referee #1, 30 Mar 2023

The authors are thankful to the reviewer for their comments and suggestions. They allowed us to improve the paper by making it clearer. After each of your comments/suggestions in quoted italics you will find the authors' s response to each comment in bold text colored on blue. Where more substantial changes were made to the manuscript, we have quoted these in bold red below. We also have made some minor corrections to spelling and wording and one citation Warnach et al., 2023 (https://egusphere.copernicus.org/preprints/2023/egusphere-2023-933/) to the final verison of this paper.

"Given the substantive dataset collected and reduced, I recommend provided overarching/broader context of the implications your data – basically, over the course of the Eruption, was the air quality adversely impacted? If so/not, to what extent, quantitatively/qualitatively – especially climate and air quality @ Earth's surface/within the boundary layer in those regions. Moreover, were their any associated human health/ecosystem/build-environment impacts associated w/ the Eruption ?"

The eruption height varied between 4 km altitude (min) and 8km altitude (max). Therefore, the eruptive emissions injected SO2 and halogen species into the free troposphere and had no major impact on air pollution at the surface even at the local scale over Sicily. Neither were there significant impacts on commercial aviation from the injection of ash. Thus, there are no impacts on human health, the ecosystem, industry, and the environment.

"I also suggest considering juxtaposing Figures 2 and 3 somehow as it would be great to see them next to each other to compare TROPOMI satellite column SO2 and BrO profiles and MOCAGE model simulated column BrO and SO2 profiles."

We agree that it is useful to be able to see Figures 2 and 3 together so that they can be compared. It is not possible to merge the two figures into one because they will be too small. However, we will try to get the editor to place the two figures next to each other in the published version.

"Also, the degree of congruence appears hard to interpret, especially for BrO as TROPOMI column profiles exhibit a background of BrO throughout the region (w/ slight variability in the region) and 6 days of Eruption while MOCAGE shows slight variability in distinct places w/ no excess BrO background (as shown in the TROPOMI BrO profiles). Atop this, is it possible to quantify the degree of congruence of the MOCAGE model results w/ TROPOMI satellite profiles ?"

We have now added Fig. S22 in the supplement of our paper to show the linear regression on a logarithmic scale of BrO from the model and the TROPOMI observations. In this figure, the red line corresponds to the linear regression and the black line represents the 1:1 line. The left column (a) shows the representation of the linear regression inside the volcanic plume and those in the right column (b), show the regression line which takes into account the values of the background outside of the plume. Each row in the panel shows a day from 25/12/2018 to 30/12/2018. We notice through these figures that the regression is not perfect and correlation are low from one

day to another for both the values of the BrO column inside the volcanic plume and the column that take into account the background.

Indeed, there is variance in both the background and plume, which is mainly caused by the statistical variation of the BrO VCD. For the 25 December, for example, the histogram of all BrO VCDs can best be described by a Gaussian distribution with a mu of $1.3x10^{11}$, i.e., almost 2 orders of magnitude lower than the sigma of $9.73x10^{12}$ molecules.cm^2. Thus, a statistical uncertainty of $2x10^{13}$ molecules.cm^2 has to be assumed, which is in line with the noise seen in figure 2 and the supplement figure S22. The mentioned systematic variations in the background are estimated to be in the order of $5x10^{12}$ molecules.cm^2, which seems to strongly impact the regression and correlation and low simulated column densities.

We realize that this is not reflected in the text and we therefore we rephrase the lines 146-150 as well as adding a short explanation of Fig S22:

"The tropospheric columns of SO_2 and BrO retrieved in the volcanic plume from the TROPOMI satellite observations around Christmas 2018 (from 25 to 30 December) obtained using a retrieval algorithm based on the DOAS method (Differential Optical Absorption Spectroscopy) (Hörmann et al., 2013; Warnach, 2022, Warnach et al., 2013) are presented in Fig. 2. The SO2 uncertainty is estimated at 35%. For BrO VCD <$4x10^{13}$ the BrO uncertainty is dominated by the statistical variation of the DOAS column retrieval, which is estimated as $2x10^{13}$ molecules.cm^2, based on Warnach et al., 2023. For higher columns the uncertainty is estimated at 35%. Furthermore, systematic biases are estimated in the order of $5x10^{12}$ molecules.cm^{-2} (Warnach et al., 2023). The systematic error component in the TROPOMI satellite observations of BrO apparently leads to relatively high and noisy background columns. For more details, we refer readers to the supplement and to Fig. S22, here it is possible to see that the relatively high systematic error significantly degrades the strength of the correlation between the model and satellite observations, particularly for the lower column densities simulated by the model. The SO_2 column for 24 December are not shown because the TROPOMI overpass was very close to the beginning of the eruption and thus only captured the plume on a few pixels."

Citation: https://doi.org/10.5194/egusphere-2023-184-RC1