Answers to the interactive comments on "Modeling study of the impact of SO_2 volcanic passive emissions on the tropospheric sulfur budget" by Claire Lamotte et al.

Comments on Pasquale Sellitto

We would like to thank Dr Pasquale Sellitto for his comments.
Our response is organised as follows. After each referee's comment (in italic black font) can be found the authors' response (in normal black font), and where needed, the changes made in the manuscript (in blue). In the revised version of the paper, only the significant changes have been coloured in blue to help identifying any new important improvement.
Also to improve the clarity of the paper and following the referees' comments, we have slightly changed the organisation of the paper by splitting section 5 into two. The new Section 5 is only devoted to the evaluation (ex-Sect. 5.1). Section 6 is on the impact of the inventory update on the species concentrations (ex-Sect. 5.2). Also, the purpose of the CARN simulation was not very clear. This simulation is only used to understand the effect of altitude of injection. This is why CARN results are only used now for the analysis of the species concentrations in the new section 6 (ex-Sect. 5.2). The manuscript has been revised accordingly.
Please note that the revised manuscript has been read and corrected by an English native speaker.

The manuscript introduces a new and more detailed volcanic emission inventory (by Simon Carn), input to MOCAGE CTM modelling, and evaluates the improvements brought in the global and regional sulphur budget with respect to older inventories using satellite observation as reference. The topics of this manuscript is of certain interest for multiple communities (atmospheric modellers, atmospheric scientists, climate scientists and volcanologists) and its worth attention. Unfortunately, I have found the following major flaws that, in my opinion, invalidate the results of this work that I think should not be published in the present form.

Major comments:

1) *In the introduction lines of Sect.3 (L150-155), it is said that SO_2 is the main volcanic effluent and is the only volcanic emission considered in this work. This is absolutely not true. The single most important volcanic effluent is not SO_2 but water vapour, with water vapour/SO_2 emitted mass ratios reaching values as large as a few hundreds.I think that many emissions and near-source volcanic processes can safely be neglected, as a first approximation, like halogen emissions and their impact on sulphate formation, transition metal contribution and other interactions of SO_2/sulphate with ash including heterogeneous chemistry; nevertheless, volcanic water vapour emissions cannot absolutely be neglected, as well as their in-plume effects on sulphate formation and SO_2 depletion. In my perspective, neglecting water vapour emissions (as said, the dominating gaseous effluent in volcanic degassing) invalidate the results of this work.*

There must have been a misunderstanding. We wrote "Among [volcanic gases], sulfur species emitted by volcanoes are mainly sulfur dioxide and hydro-sulfuric acid in much lower quantity". Here, we were not trying to say that sulfur dioxide is the main effluent but to explain that sulfur dioxide is the main effluent for sulfur species. The sentence has been changed in the revised manuscript to make it clearer.
Indeed, water vapour is the most important volcanic effluent and could be taken into account in volcanic emissions. We agree with these statements. There is an effect of volcanic water vapour on the sulfate formation within plumes taking place close in time and space to the emission/eruption.
In our study, we do not take into account this effect as in similar previous studies at the global scale (e.g. the Sheng et al. paper you suggested to cite, updated in Feinberg et al. (2019)). The reasons are that we use a global chemistry-transport model with a resolution of 1° x 1°and run over one year. With this resolution, if we were to include water vapour volcanic emissions and associated sulfate production, the water vapour emitted would be diluted into the grid box (about 100km x 100km) such that its effect on the acceleration of sulfate formation would be negligible. More importantly, the effect of the water emitted by the volcanoes is mainly important in the first stages of the plumes in the vicinity of volcanoes, small scale processes that we cannot and do not intend to represent in our global model at 1° x 1°resolution. The aim of the paper is not to focus on small time and space scales close to the volcanic emissions but to assess the impact at the global scale of sulfur emissions of all volcanoes on the sulfur budget. Note also that MOCAGE simulations use meteorological analyses using data assimilation of water vapour information. Therefore, some of the volcanic water vapour can possibly be taken into account in the meteorological analyses via data assimilation of water-related observations.

2) *I agree with Referee #1 on the fact that ACSAF GOME-2 retrievals are not a good choice for the validation of the MOCAGE simulations. I'd also mention that, differently to what said at L287, GOME-2 data are not completely independent on the OMI and TOMS input data to your inventories: the instruments operate in the UV spectral range and use similar spectral ranges and SO_2 absorption structures for the retrieval. Why not using infrared instruments as IASI, instead?*

We agree with the reviews that GOME-2 MetopA SO_2 dataset shows unrealistic features in some regions. Although we had applied filtering on these data, it was not enough to remove all the noise and artefacts.
Concerning IR instruments, such as IASI, they have the advantage to be fully independent of OMI since using different wavelengths. Moreover they have the ability to retrieve SO_2 columns at high latitudes in winter or at night. However, IASI is mainly sensitive to SO_2 in the mid and upper troposphere but not very much in the planetary boundary layer (or under 5 km). Even with more sophisticated algorithms designed to extract information below 5 km, the estimates of SO_2 columns are shown to be less accurate in the lower troposphere and to underestimate small emissions sources [Carboni et al (2012), Taylor et al (2018)]. This is why in the literature, IASI was mainly used to study eruptive events, emitting at higher altitudes [Clarisse et al (2008, 2014), Carboni et al (2016, 2019)]. Therefore, we think that IASI measurements are not suitable for the model evaluation for the year 2013, since 2013 has very few eruptive events and thus volcanic emissions are mostly emitted below 5km. Still, it would be interesting to use IASI SO_2 columns for a year with more and higher eruptions.
Concerning the validation strategy in the paper, we no longer used GOME-2 SO_2 columns but OMI SO_2 columns as suggested by referee 1. Even if OMI has been used for in the Carn's inventory, it has the finest resolution and it is the most accurate instrument in 2013 to retrieve SO_2 total columns over passively emitted volcanoes which altitudes are generally around 2-3 km. We also changed the approach chosen for the statistical evaluation based on the analysis of the literature. In satellite derived products, the relative uncertainties on SO_2 columns are large where the SO_2 signal is low, in particular for background SO_2 conditions. This is why in the literature, the SO_2 satellite comparisons or model evaluations focus on specific areas close to SO_2 sources [*e.g.* He et al. (2012), Fioletov et al. (2013), Wang and Wang (2020)]. Similarly to these studies, our new strategy is to perform the model evaluation only in the vicinity of the volcanic sources. The comparison between the model and OMI SO_2 columns clearly show an improvement of the model performances in CARNALTI simulation. Section 4.2 "Observations used for the evaluation of the simulations" and 5.1 "Evaluation of the simulations" were rewritten to explain our new model evaluation strategy and associated results. Further details are explained in the answer to Referee #1.

3) *Also, the choice of MODIS AOD is quite debatable strategy. MODIS AOD is linked to all aerosols, not only sulphates. How do you separate sulphate aerosols from the other aerosol typologies/composition/sources? For example, in "Region 3 (Mediterranean)" dust is, on average, overwhelmingly dominant with respect to sulphate aerosols. How can you check the improvement of volcanic SO_2 sources in such an environment, due to the expected small sulphate signal?*

We are aware that MODIS AODs include all aerosols but satellite observations of sulfate aerosols only at the global scale are not available. MODIS AOD is an alternative allowing us to make an indirect evaluation since it takes into account all types of aerosols, sulfate included. Between the different simulations, only SO_2 volcanic emissions are modified. So, the changes between the model simulations with respect to MODIS AODs come only from sulfate aerosols.
Regarding dusts, they contribute largely to AODs seasonally in some regions and can partially/totally hide the sulfate contribution to AODs. Still note that dusts also help forming sulfate aerosols. In the paper, we had highlighted that in some regions (mostly polluted areas), those changes are very small because hidden by anthropogenic emissions. This is further discussed in the revised version.
Note also that in the validation process, with MODIS or another instrument, we are interested in the differences between REF and CARNALTI simulations. In general, the validation of the AODs show a decreasing FGE and an increasing correlation between REF and CARNALTI. We consider this result satisfying.

4) *In addition, how to interpret the results of the comparisons in Tables 3 and 4? Am I wrong to say that observations and simulations compare very weakly? This is also the case if looking at the (necessary in the main text) Figure S1. The simulations and the observations seem to not describing the same SO_2 fields. Results for aerosols compare better but, in my opinion, only because the aerosol fields are dominated by other aerosols (and MODIS is more sensitive to higher altitudes aerosols than boundary layer aerosols, so again probably large dust plume lofted by convection).*

Tables 3 and 4 are results from the comparison with GOME-2 Metop A. Since we now use OMI instead of GOME-2, the part of the paper concerning the SO_2 evaluation has been fully revised (see answer to Major Comment 2).

Concerning aerosols validation, Fig. 11b clearly shows that volcanic emissions can have a strong contribution in aerosol sulfate concentrations. Even over polluted area, such as the Mediterranean Sea, sulfate aerosols from volcanic emissions contribute to about 10-20%. This is not negligible.

5)*In general, the manuscript is poorly written and needs a thorough linguistic review. The description of the scientific context is quite approximative and a lot of key references are lacking – please see specific comments in the following.*

The manuscript has been revised and corrected following the suggestion of all reviewers and fully checked by a native English speaker. Several references have been added in the revised manuscript. We preferentially chose those focusing on global studies rather than case studies, the latter being of lower relevance for the aims of the paper.

Technical comments:

1) *L1: "The contribution. . . ": what contribution?*
Here, we talk about the contribution of volcanic sulfur emissions, with respect to other sulfur emissions. We emphasise that volcanic emissions allow more sulfur species to remain into the troposphere, and especially sulfate aerosols. But this sentence was not appropriate here $\longrightarrow$ deleted.

2) *L9: "negliable" → "negligible"* $\longrightarrow$ corrected

3) *L20: Here and all the following discussions (including comparisons with your results): recent assessments of sulphur budget should be discussed here, like (for volcanically quiescent conditions, so of large interest for our study): Sheng et al (2015).*

Thank you for the suggestion of the Sheng et al. (2015)'s reference which was updated with improved model simulations in Feinberg et al. (2019). Even if these two studies focus on the stratosphere, they provide a sulfur budget estimate also for the troposphere. Comparisons to Sheng et al. (2015) and Feinberg et al. (2019) have been included in the revised manuscript.

4) *L-20: Also, for climate impacts, this should be cited and possibly discussed: Kremser et al. (2016)*
We have included the reference as suggested but no addition discussion since the climate impact of sulfur is out of the scope of our paper.

5) *L22: "variation of climate": You mean "climate forcing"? (in this case, please specify that you're not talking about SO_2 but sulphate aerosols)* $\longrightarrow$ We meant sulfate aerosols climate forcing. This has been corrected.

6) *L23-24: "SO_2 emissions had become a major concern in environmental policies, leading to strong reductions in anthropogenic emissions in recent decades": Not everywhere. Please differentiate geographically between decreasing, stationary and increasing emissions regions and add a reference.*

Indeed, changes in anthropogenic SO_2 emissions are not similar worldwide. Details were added in the revised manuscript as follows:
In some regions of the world, these policies led to strong reductions in anthropogenic SO_2 emissions in the recent decades [Fioletov et al. (2016), Krotkov et al. (2016), Aas et al. (2019)]. Over North America and Europe, emissions strongly decreased between 2005 and 2015. In the East Asian region, the decrease only happened after 2010 [Sun et al. (2018)]. On the contrary, over India, emissions strongly increased. And over other large SO_2-emitting regions (Mexico, South Africa, Russia or Middle East), they remained stable since 2000. However, the decrease in anthropogenic SO_2 emissions over Europe and North America was sufficient to induce an overall decrease at the global scale.

7) *L23-24: "Thus, the relative proportion of volcanoes in the total sulfur emission sources tends to increase.": Due to different regional trends of anthropogenic emissions, this statement sounds just arbitrary (unless you have specific references that I don't know).*

Despite the increase of anthropogenic SO2 emissions in India, the statement about the decrease in anthropogenic SO2 emissions at the global scale is true. With respect to anthropogenic emissions, we think that it is acceptable to say that at the global scale, the relative proportion of volcanic emissions in the total sulfur emission sources tends to increase.

8) *L26-27: "is greater in altitude": you might mean: "increases with altitude"* $\longrightarrow$ corrected

9) *L27-28: "Thus, we now. . . emissions": not clear, please rephrase* $\longrightarrow$. Removed

10) *L27: "longer": "for longer time periods"?* $\longrightarrow$ corrected

11) *L29: "these variations": which variations?* $\longrightarrow$ "these" unnecessary and deleted to clarify.

12) *L44-45: Please change the phrasing here: there are very few "easy-to-access" volcanoes (Masaya can be mentioned, maybe), while normally the internal processes themselves build "uneasy-to-access" morphological structures for volcanoes.* $\longrightarrow$ rephrased

13) *L51: "information on injection altitude is available". The information on the altitude is still very limited. These are observing systems that have a few units of Degrees of Freedom in vertical profile observations of SO_2, mostly between 1 and 2.0-2.5, so not allowing for detailed altitude information. Please mention this in the text and smooth this statement.*

We agree that emissions inventory, and especially those built by satellite observations, should not be considered as the absolute truth, because there are uncertainties in the retrievals, both for the SO_2 quantities and the plume height. Moreover, in MOCAGE, there are only 47 vertical levels. Into the free troposphere, each levels are separated by at least 200-300 m and volcanic emissions are injected on the closest vertical level of the plume altitude. Therefore, this adds more uncertainties to the plume height. We added a statement in Section 3.2 "New volcanic sulfur inventory" as follows:
Note that depending on the instrument used, the retrieval of the plume altitude can differ. Therefore, there are uncertainties on the altitude information provided by this inventory.

14) *L56: what do you mean with "more numerous and qualitative data"?*

This sentence means that, with the improvement in the retrieval of SO_2 emissions by satellites, Carn et al (2016,2017) inventory includes more data over more volcanoes and with a higher quality, with respect to Andres & Kasgnoc (1998) inventory $\longrightarrow$ rephrased as follows:
Carn et al (2016,2017) sought to compile all those new higher quality data, compared to Andres & Kasgnoc (1998), in order to provide a more representative inventory of volcanic SO_2 emissions.

15) *L63-65: "In contrast, few studies focus on the impact on tropospheric composition including air quality, with the exception of case studies of volcanic eruptions...": This is not true. Please look at the following papers of my research group, that aimed at the impact of volcanic activity, including passive degassing of selected volcanoes, on the tropospheric composition and air quality. Please correct the wrong statement and cite the previous work mentioned above.*

There is a misunderstanding here. In this sentence, "In contrast" was a reference to studies at the global scale only, similar to ours. Some studies, such as yours, analysed the impact of volcanic degassing (and not only eruptions), but at the regional scale. Thank you for the references. We have added more prescise information in the revised version.
At the global scale, numerous studies aim at the assessment of the dispersion of sulfate aerosols and the subsequent radiative forcing [Graf et al. (1997,1998), Gasso et al. (2008), Ge et al. (2016)]. In contrast, regarding their impact on tropospheric composition, including air quality, several case studies at the regional scale have been analysed [e.g. Colette et al. (2010), Schmidt et al. (2015), Boichu et al. (2016,2019), Sellitto et al. (2017)] but very few studies at the global scale have been conducted [Chin et al. (1996), Sheng et al. (2015), Feinberg et al. (2019].

16) *Section 2.5: What about the vertical transport, which can pose problems for the modelling of confined plumes, like volcanic plumes, and is discussed in the following paper [Lachantre et al. (2020)]?*

Lachatre et al (2020) is a good reference about the vertical diffusion of plumes linked to the modeling of vertical transport. But in our analysis at the global scale, we do not focus on the study of individual volcanic plumes but on the global fate of the sulfur volcanic emissions and in particular on their impact once dispersed. As in all global models, MOCAGE description of volcanic plumes is limited by both the vertical (at least 200-300m in the free troposphere) and the horizontal (1° x 1°) resolutions not allowing the detailed modeling of individual plumes.

18) *L165: "calm": What do you mean with "calm"? "Non-eruptive"?*
"Calm eruptive conditions" do not mean "non-eruptive", but with small eruptions and not strong eruptions. The term used in Andres & Kasgnoc (1998) is "quiet" $\longrightarrow$ corrected

19) *L167: the reference to molecular structure sounds strange here. You might want to say that "SO_2 and ozone have absorption bands at overlapping spectral regions" (which is linked to molecular structure) or something like.*

The reference to the similar molecular structure of ozone and sulfur dioxide was difficult to understand. The two species have overlapping UV absorption bands (between 300-340 nm). Therefore, TOMS' measurement of SO_2 is tangled to O_3. Krueger et al (1995) explained this phenomenon as follow. "Typically, the amount of sulfur dioxide in the region of the atmosphere that affects TOMS-measured radiances (above the boundary

layer) is too small to cause significant absorption. However, a volcanic eruption can produce enough SO2 in a localized region to produce UV absorption comparable to or even exceeding the ozone absorption at the shortest two TOMS wavelengths. In such cases the present TOMS algorithm incorrectly interprets SO2 as enhanced ozone. The problem is to discriminate between sulfur dioxide and ozone.". Thus, an algorithm is needed to discriminate ozone from sulfur dioxide measurements. This level of details is not necessary. The sentence has been deleted.

20) *L181: "...as one of the largest passive emitters": Clumsy phrasing. Please rephrase.* $\longrightarrow$ rephrased as follows:
Etna in Sicily, Kilauea and the Kilauea Rift Zone in Hawaii, which are known as being among the largest emitters of SO_2.

21) *L181-182: Please add details on the sources of these flux information.*

The original statement in Andres & Kasgnoc (1998) is :"For three sites, however, personal communications supplanted the average. These personal communications relied upon published and unpublished data for Etna, Kilauea and Kilauea East Rift Zone." No other information is available. Therefore, we cannot add details in the paper.

22) *L183-184: This is very unclear. Please clarify.*

We agree that the explanation we gave in the paper on sporadic eruptions can be confusing. We could have explained it as follows: "With regard to sporadic eruptions that are considered for 25 volcanoes, Andres & Kasgnoc (1998) use the maximum flux reported during the period and assume an average of 7 eruptions per year, each lasting one day. From this, sporadic eruptions account for less than 1 % of the total annual emissions in their inventory". However, in the next paragraph, we explain that eruptions are not taken into account in the model. Therefore, this level of details is not needed $\longrightarrow$ The sentence has been deleted.

23) *L188-189: You mean that volcanic SO_2 is emitted at the surface (including orography)? Is orography "smoothed" by the average in-grid topology? This aspect is very important e.g. for Etna. Even in case of passive degassing, its emissions are released at, at least, 3000 m altitude and episodic eruptions can reach, for Etna and Kilauea, quite higher altitudes.*

The model surface altitude corresponds to the model orography which is calculated as the average in-grid topography. This means that in the previous version of MOCAGE, the volcanic emissions which are emitted at the surface are mostly under the actual volcano altitude. This is now clearly mentioned in the revised paper.

24) *Section 3.2: there are many repetitions. In general, all the paper should be condensed and repetitions should be suppressed.* $\longrightarrow$ corrected

Section 3.2 has been changed according to referees' comments and overall reduced in the revised version.
25) *L224-226: "We implemented...emissions": Why this parameterisation is not described in details here? How it compares to established parameterisations like the one of Mastin et al (2014)?*

For each eruption, we use the altitude of the volcano and the height of the eruption given in Carn et al. (2016) inventory. This information is derived from the analysis of nadir UV and IR satellite observations. Therefore, we do not need to make an estimation of the eruption height by the use of a parameterizations like the one proposed by Mastin et al. (2014). Still, in the model, we have to distribute vertically the mass of SO_2 given in the inventory. In MOCAGE, we distribute the eruption emission mass from the model level of the volcano altitude to the model level of the plume top height, following an "umbrella" profile similar to that used in other models (Freitas et al. 2011 in CCATT-BRAMS and Stuefer et al. 2013 in WRF-Chem). In practice, the plume follows an almost linear profile with increasing altitude from the volcano vent and then opens into a parabola containing 75 % of the gases in mass into the top third of the plume. This paragraph has been re-written in the revised version in order to be clearer.

26) *L239: "Finally, the availability of emission heights in this inventory gives a better description of the emission". At this point I think it is necessary to discuss the limitations in the vertical characterisation of volcanic emissions in the new inventory and the satellite observations used to build it, so to not oversell your new simulations.*

We fully agree and we have added a sentence on the uncertainties of the inventory and satellite observations in this section. We also added a paragraph on this subject in the conclusion.

27) *Figure 1 and most figures: Please use larger text and labels.* $\longrightarrow$ corrected

28) *L269: "lowest eruptive. . .negligible in 2013": How much this is "low"? Is it really negligible? How do you qualify this as "negligible"?*

This information is given just after "the total 2013 annual emissions in Carn et al (2016, 2017) inventory amount to 23.7 Tg of SO_2 (or 11.8 Tg S), with 23.5 Tg of passive degassing SO_2 and 0.2 Tg of eruptive emission (< 1 % of the total amount of volcanic SO_2 emission)". This part of the paragraph was changed in order to make this clearer.

29) *L272: reference to summer and winter: Please correct to "northern hemisphere summer/winter" and adapt the discussion.* $\longrightarrow$ rephrased.

30) *L284-285: "Due" and "since" in the same sentence is quite clumsy. Please rephrase.* $\longrightarrow$ rephrased as follows:
The target chemical species that we evaluate are SO_2 and aerosols, since SO_2 is the precursor of sulfate aerosols.

31) *Section 4: see Major Comments 2-3* $\longrightarrow$ Taken into account. This section has been revised (see answers to comments 2-3)

32) *Section 4.2.2 title: "MODIS Aerosol Optical Depth"* $\longrightarrow$ corrected

33) *L349: Please check altitude of Mount Etna, this is not the right altitude.*

We agree that the altitude of Mount Etna is about 3330 m but the altitude provided in Carn et al. (2017) inventory is 2711 m. No information is given in the documentation/publication why this altitude is lower in the inventory. It possibly accounts for passive emissions from volcano flanks or a mistake. Nevertheless, the aim of the paper is to implement a new volcanic emissions inventory and to evaluate it as a whole in the model, even if there are possible uncertainties in the altitude of the volcanoes or other parameters.

34) *Section 5: see Major Comment 4* $\longrightarrow$ taken into account

35) *Section 5: It looks like some of the Figures in the Supplements are needed here in the main text, e.g. S1* $\longrightarrow$ with the new validation strategy, now not necessary, deleted.

36) *L430: "(industries...': and dust, of course* $\longrightarrow$ added

37) *Figure 8: This figure would be largely more useful with an altitude vertical axis (instead of pressure).* $\longrightarrow$ We have added the altitude axis and enhanced the labels for the pressure axis.

38) *L517: "This corresponds...eruption": This is quite straightforward interpretation of these results, but it is important to stress the fact that 2013 is not a "normal" year as even a small number of explosive volcanic eruptions can change the vertical distributions of Figure 8 at the global scale. This has to be discussed and the limits of your simulation (a" predominantly passive degassing" year) must be clearly stated.*

There is a misunderstanding here because this sentence is a comment on Graf et al (1997) results. We changed the paragraph as follows:
For volcanic sulfate, the maximum is between 850 and 450 hPa but four times smaller than for other sources and without any specific peak associated to passive degassing or eruptive emissions. These results are different from Graf et al. (1997), which shows that the vertical distribution of volcanic sulfate aerosols is comparable to anthropogenic and biomass burning sulfate and is even dominant between 800 and 300 hPa (the altitude of volcanic emissions, mainly from eruption). This difference between our study and Graf et al. (1997) can be explained by the quantity of SO_2 emitted by eruptions. In 2013, only a few eruptive events occurred while almost 30% of volcanic emissions in Graf et al. (1997) are eruptive. Therefore, with a greater amount of volcanic emissions injected at higher altitude in Graf et al. (1997), the potential to form sulfate aerosols is greater than in our study. This can explain the greater efficiency of 2.63 in the tropospheric sulfate burden in Graf et al. (1997) compared to 1.89 in our study.
In Fig. 8, we had not clearly discussed about the impact of eruptive emissions on the vertical distribution. We added this statement in the revised paper:
[SO2 vertical profile] There is no contribution below 950 hPa but there are three maxima above; one at 850 hPa (about 1500 m) due mostly to passive degassing, another around 680 hPa (about 3300 m) due to passive degassing from high-altitude volcanoes and eruptions, and the last one around 450 hPa (about 6000 m) due to high-altitude eruptions. It is noteworthy that even with few eruptive events during the year 2013, the volcanic SO_2 vertical distribution is affected by them.

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