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 Anonymous Referee #2

We would like to thank the Anonymous Referee #2 for its comments that helped improving the paper.
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 and that we have added co-authors to the paper that contributed to the responses to the referees and to the revised version.

The paper by Lamotte et al. studies the effect of SO_2 volcanic degassing emissions on the tropospheric sulfur budget, using the MOCAGE global CTM implemented with new volcanic emission inventory of Carn et al. (2016,2017). By model sensitivity tests, tropospheric SO_2, sulfate and AODs simulated with new and old emission inventories for the year 2013 are compared and validated against SO_2 GOME-2 and AOD MODIS satellite data sets. The results show that the new inventory (CARNALTI) is the best with reference to satellite observations. Such kind of study is interesting and should provide important information for understanding the sulfur global budget. On the other hand, in the opinion of this referee, the manuscript still needs improving a lot before acceptance for publication.

1A) *I agree with Referee #1 and Dr. Pasquale Sellitto that other observational data should be used for the validation of the model simulations.*

We agree with the reviewer that the Metop-A GOME-2 SO_2 columns presented show unrealistic features in some regions. Not being experts on satellite observations, we had chosen for the model evaluation to use GOME-2 MetopA SO_2 columns from DLR provided by ACSAF (ex- O3F-SAF) because those data provide an independent measurement of SO_2 with respect to OMI (used in the volcanic emission inventory). Indeed, these data present artefacts and noise. Although we had applied filtering, this was not enough to remove all the unrealistic features. This is probably the reason why these data were mainly used in the literature not at the global scale but on case studies at the regional and local scales [Rix et al (2009,2012), Koukouli et al (2015)], and to detect very large emission sources [Fioletov et al (2013)]. Note that we also investigated the use of GOME-2 MetopB SO_2 columns from DLR by ACSAF (ex- O3F-SAF) but the results showed similar unrealistic features in some regions as in GOME-2 MetopB SO_2 columns. Concerning IR instruments, such as IASI, they are mainly sensitive above 5km which is too high for our study focused on passive emissions.
This has lead us to change our evaluation strategy. As suggested by Referee #1, we choose in the revised version to use OMI SO_2 columns data for the model evaluation. We also changed the approach chosen for the statistical evaluation based on the analysis of the literature. As for all 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. For each volcano, we select 9 model grid points (representing a square of 3°longitude x 3°latitude) with the middle point being where the volcano is located. 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.

1B) *In addition, I am very concerned about the effects of the volcanic emission heights on the simulated results.*

Tables 1 and 2 present different simulation scenarios, but I missed the detailed information on emission heights of the new inventory of Carn et al. (2016,2017). As I understand, the emission heights are provided in the work of Carn et al. (2016,2017), but for the sensitivity test, all volcanic emissions (eruptive and passive) were arbitrarily forced to the model surface for the scenario CARN.

We were not clear enough, but your understanding was right. The aim of the CARN simulation is to make a sensitivity run in support of the analysis of the differences between REF and CARNALTI. The major differences between Andres and Kasgnoc (1998) and Carn et al (2016,2017) are the updated quantities of volcanic emissions and the information on plume altitude. CARN simulation allows us to distinguish between the impact of the height of emission and of the quantity of SO_2 emitted. This is why in the revised manuscript, CARN results are only used for the analysis of the species concentrations in the new section 6 (ex-Sect. 5.2) The paragraph describing the different simulations was rewritten as follows:
The first simulation, named REF, takes into account the previous volcanic inventory [from Andres and Kasgnoc (1998)] with the injection at the model surface. The second simulation, named CARNALTI, uses the updated volcanic inventory [from Carn et al. (2016, 2017)] and the new configuration to inject volcanic emissions from the volcano altitude as described in Section 3.2. By comparing REF and CARNALTI runs, we can analyse the changes brought by the the updated volcanic emission inventory with respect to the previous one. These two simulations are evaluated in Section 5 and the associated global distribution of sulfur species is compared in Section 6.
In order to distinguish between the impact of the height of emission and of the quantity of SO_2 emitted, another simulation, named CARN is run and used for the analysis of the differences between REF and CARNALTI global distribution of sulfur species. Volcanic emissions are from Carn et al. (2016, 2017), like in CARNALTI but they are injected at the model surface, like in REF.

1C) *While the SO_2 tropospheric columns are compared, can the vertical distributions of SO_2, sulfate or aerosols be better simulated using CARNALTI than with CARN?*

With different altitudes of emission between CARN and CARNALTI simulations, we expect higher aerosol content at altitude with CARNALTI simulation (as explained in section 5.2 "By injecting volcanic emission in altitude with the new configuration in the simulation CARNALTI, less sulfur species remain at the surface and therefore aerosols are spread further from the volcanoes (see Fig 7b)"). Moreover, the vertical variability of winds also induces differences in the horizontal aerosol distribution. In CARNALTI, SO_2 volcanic emissions are more realistically distributed vertically leading to an expected improvement of the overall vertical and horizontal distributions of SO_2.

2) *2.3 Emissions (L 109-114): The emission inventories (MACCity and GFAS) are for the years before 2010, earlier than the simulated year 2013. Can this affect the comparing results?*

As written in the paper, anthropogenic emissions from MACCity inventory and biogenic emissions from MEGAN-MACC inventory are representative of the year 2010. However, the differences between 2010 and 2013 emissions are not very important (see Figure 1). At the global scale, SO_2 emissions are only about 1% higher than in 2013. Locally, it represents only a reduction of 8% over oceans/seas between 2010 and 2013, and an increase of 7% over North Africa. Therefore, the expected impact of the use of the 2010 emissions instead of 2013 is low. We have added a sentence in the revised manuscript stating that the differences between 2013 and 2010 are small.
For GFAS products, it is a database available since 2012 with daily biomass burning emissions for each year since then. In this study, we used the daily GFAS data for 2013. We make it clearer in the revised version of the paper.

3A) *2.4.1 Gaseous species (L 123-125): Two schemes, RACM and REPROBUS, are used for tropospheric chemistry and stratospheric chemistry, respectively. How the model grid cells are distinguished between the troposphere and the stratosphere so that only one of them is applied?*

The chemical scheme used in MOCAGE is a merge of RACM and REPROBUS so that no distinction between the troposhere and the stratosphere is needed. This means that all chemical species are defined at all gridpoints in MOCAGE. The following changes are made in the manuscript.
The MOCAGE chemical scheme is named RACMOBUS. It merges two chemical schemes representing the tropospheric and stratospheric chemistry. The first one, the Regional Atmospheric Chemistry Mechanism (RACM) (Stockwell et al. 1997), completed with the sulfur cycle [details in Guth et al. (2016)], represents tropospheric species and reactions. The second one, REactive Processes Ruling the Ozone BUdget in the Stratosphere (REPROBUS), provides the additional chemistry species and reactions relevant for the stratosphere, in particular long-lived ozone depleting substances (Lefevre et al. 1994).

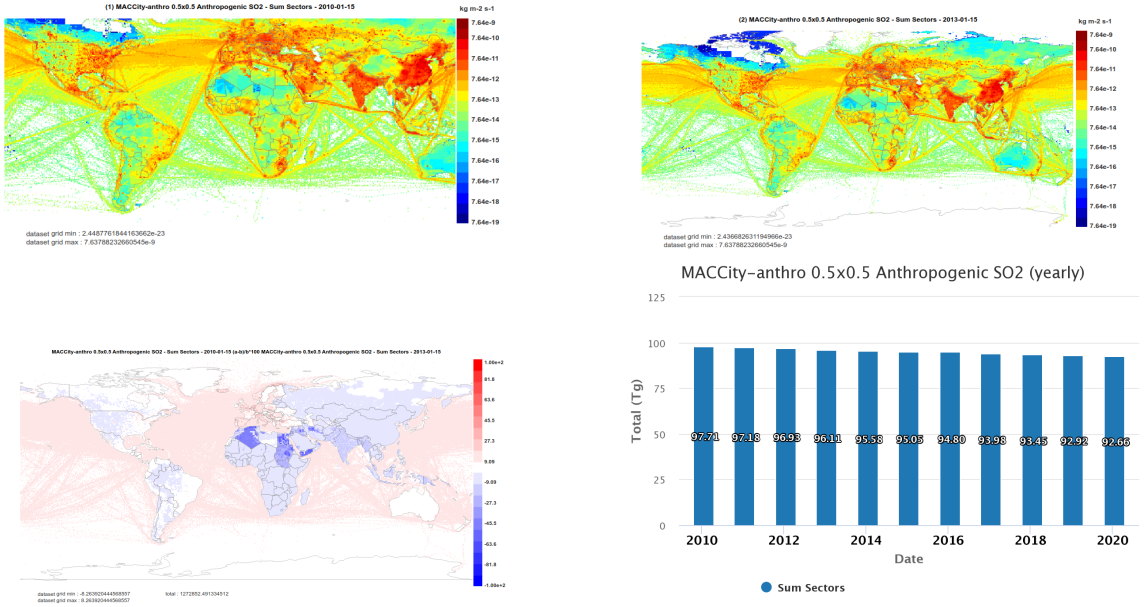

Figure 1: SO_2 total emissions (all sectors) from MACCity inventory for the year (upper-left) 2010 and (upper-right) 2013. (lower-left) Relative difference between 2010 and 2013 SO_2 emissions in MACCity. (lover-right) Time series of the total annual SO_2 emission in MACCity inventory from 2010 to 2020.

3B) *How are stratospheric tracers (e.g., CFCs and OCS) and tropospheric tracers (e.g., NMVOCs) treated in the model grid cells?*

Long lived species relevant for the stratosphere (e.g., CFCs) are fixed at the surface, similarly to many other global models. Tropospheric VOCs undergo chemical processing in the troposphere leading to negligible concentrations reaching the stratosphere as expected. Still, there is one exception of species that is represented with two different model variables. It is for H_2SO_4 for which a climatology is used in the stratosphere and which is treated as a 'normal' species in the troposphere In the paper, we analyse tropospheric sulfur only.

3C) *Can TUV calculate the photodissociation rates of stratospheric chemical tracers?*

Yes, TUV model can calculate photodissociation rates for both the troposphere and stratosphere. This detail is now explained in the revised manuscript.

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