Reply to general comments of RC1:

We thank the reviewer for the time to review our manuscript, and especially the very detailed and constructive feedback. We reply to the general comments before the end of the discussion phase, while we will present a detailed reply on the specific comments once the discussion phase is concluded. We report the comments (grey, bold) along with our replies (blue).

The submitted manuscript contains the description of a new submodule within MESSy. The submodule is described in detail in section 2. Section 3 provides a description of the model setup for the validation of the submodel. For model validation the 2011 Nabro and 2018 Kilauea SO_2 emission plumes were evaluated with different sensitivity simulations and with a fine-resolution simulation, respectively. In chapter 4 the explosive volcanic SO_2 injections of the time period 2007 to 2011 was simulated based on a 3-D observational emission dataset which was improved based on the findings of chapter 3.

The submitted manuscript describes a new submodule for emission of volcanic substances and provides some validation for the evolution of SO_2 plumes after volcanic eruptions. However, in my point of view, the manuscript has some substantial flaws, which I explain below and which need to be addressed. Even though topic wise the manuscript would be a good fit for publication in GMD, I do not think that quality wise this manuscript deserves publication in this journal.

We thank the reviewer for the assessment of our manuscript. We agree that there are some mentioned shortcomings that have to be addressed as pointed out, however, we disagree on the overall evaluation. We will discuss the general comments in detail below.

The main environmental impact of large explosive volcanic eruption on a global scale (heterogeneous chemistry, cooling of the surface, local warming of the stratosphere) are effects from sulfuric acid aerosols and not from SO_2. The manuscript almost entirely focuses on the simulation of SO_2 plumes and thus, a crucial aspect of volcanic emission plumes is missing. I know that for the accurate simulation of aerosol burden and their effects it is important to accurately simulate the SO2 plume in the first place, however, if this was the goal of this study this needs to be motivated and explained in the introduction.

We agree with the reviewer that the main environmental impact of volcanic eruptions comes from sulfuric acid aerosols. However, as the reviewer pointed out, these sulfuric acid aerosols are a result of volcanic SO_2 emissions. We also state this twice in the introduction: "Moreover, SO_2 undergoes oxidation to form sulfuric acid H_2SO_4, which rapidly converts to the aerosol phase forming sulfate under most atmospheric conditions." and "When volcanic SO_2 emissions reach the stratosphere, the subsequently formed sulfate aerosols enhance the stratospheric aerosol burden and are distributed widely across the globe". Therefore, for the evaluation of the EVER submodel, SO_2 is the correct tracer that needs to be investigated and evaluated. Nevertheless, the effects of sulfuric acid aerosol are described in the introduction, as they have the most important impact, and they result from volcanic SO_2 emissions.

Indeed, the goal of the study is to present a new submodel that handles emissions of gaseous and aerosol species, and the accurate simulation of volcanic SO_2. The subsequent formation of sulfuric acid and aerosols is a consequence of chemistry, thermodynamics and microphysics.

This is handled by the other submodels present in the MESSy model system (e.g. MECCA and GMXe), which have been largely evaluated elsewhere and therefore are not subject of this paper. We also state this in the introduction: "Our primary aim is to provide recommendations regarding the implementation of stratospheric SO_2 injections from volcanic eruptions within the MESSy framework ...", "We evaluate the EVER submodel using SO_2 emissions from the 2011 Nabro explosive volcanic eruption ..." and "Based on the stratospheric volcanic SO_2 emission inventory developed by Schallock et al. (2023), we establish a default setup for the EVER submodel ...". We want to make this clearer in the abstract and the introduction.

Regarding the comment "a crucial aspect of volcanic emission plumes is missing": We do not agree on that. The EVER submodel is only responsible for the emission of the initial plume species. Sulfuric acid aerosols are not part of the "volcanic emission plume". They form in the evolution of the plume. Standard aerosol simulations with EMAC (or MESSy in general) already included the chemistry, thermodynamics and microphysics of sulfuric acid aerosol (see for example Brühl et al., 2018; Schallock et al., 2023), however it was missing a standard setup for including the initial SO_2 emissions, which we wanted to address.

As we agree, that the long-term SO_2 mixing ratios (Sect. 3.3.2) and the resulting aerosol evaluated later depend on the SO_2 removal and microphysics, we will talk about their handling in more detail in a revised version (see also comments below), but this is a more technical comment in our opinion. Moreover, the chemical mechanism, including H_2SO_4 formation, is already provided as supplement, which we refer to in the manuscript.

To me many important aspects of a paper are not sufficiently explained: What is the novelty of this study? What can the model do what other models can't do? Why is it important to simulate the emissions of SO_2 accurately? Why do we need yet another model which is capable of simulating volcanic injections? What have other models shown and how does your model compare to other models?

This manuscript is a documentation of a new submodel, along with its evaluation and the development of a default setup. It is not a study, aiming to provide new results on volcanic eruptions. This is well within the scope of GMD papers. From the "Aims and Scope" of GMD papers (`https://www.geoscientific-model-development.net/about/aims_and_scope.html`) amongst others:

- "geoscientific model descriptions, from statistical models to box models to GCMs"
- "development and technical papers, describing developments such as new parameterizations or technical aspects of running models such as the reproducibility of results"

We chose the manuscript type "Development and technical papers" (from `https://www.geoscientific-model-development.net/about/manuscript_types.html`): "These papers describe technical developments relating to model improvements such as the speed or accuracy of numerical integration schemes as well as new parameterisations for processes represented in modules. [...] Development and technical papers usually include a significant amount of evaluation against standard benchmarks, observations, and/or other model output as appropriate."

We think that most of the raised questions are already sufficiently answered within the scope of a GMD paper.

- **"What is the novelty of this study?"** In MESSy, we so far only had a more or less generic submodel (TREXP, Jöckel et al., 2010) for point or column like emissions in the atmosphere. This submodel, however, was made for different purposes, and missing features which are particularly required for emissions from volcanic eruptions (e.g. the possibility to specify vertical emission profile shapes or primary aerosol emissions as needed for volcanic ash). To avoid increasing the complexity of TREXP, we decided to implement EVER. We describe this in lines 125ff. In previous studies a lot of manual work was necessary to consider explosive volcanoes and other injections into the stratosphere, and there was no standard setup covering stratospheric volcanic eruptions.

- **"What can the model do what other models can't do?"** Indeed, other models can also handle volcanic eruptions; however, in the MESSy model system, it was not possible so far. So, we don't see any contradiction here. We present our new submodel and evaluate it using observations. The implementation differs from other models being specific to the MESSy interface, although we can refer more to other model developments.
- **"Why is it important to simulate the emissions of SO_2 accurately?"** Details about the importance of sulfuric acid aerosols resulting from volcanic SO_2 emissions are listed in the introduction. This implies the importance of accurately simulating SO_2 emissions.
- **"Why do we need yet another model which is capable of simulating volcanic injections?"** As there was none in MESSy, and other available models were not sufficient for our needs, this paper described a further development of the code. We will make it clearer in the introduction, why we chose an own implementation, and thank the reviewer for this suggestion. We will also add a more detailed part in the introduction, where we discuss available volcanic models.
- **"What have other models shown and how does your model compare to other models?"** While we mostly focused on comparison with observations, we will highlight the differences and similarities to other model studies also in a revised version.

To avoid further confusion, we want to provide a clearer description of the nature and the goal of the study in the abstract and the introduction.

I think the manuscript does not provide/discuss a substantial part of literature relevant for the simulation of volcanic emission plumes. There have been many studies, which highlight important aspects discussed in this manuscript, which are not even cited (e.g. Brodovsky et al. 2021 or Quaglia et al. 2023).

We thank the reviewer for the comment. We will include more model literature in a revised version, and agree with the reviewer that this literature is crucial to discuss.

Usually, the 1991 Mt. Pinatubo eruption is used as a basis for model validation (e.g., Quaglia et al. 2023, which even includes EMAC simulations). Why did you not perform a simulation of Mt. Pinatubo to validate your model? Probably because this has been done extensively already, but it would allow comparing also with other models.

We chose the Nabro volcano, as there are extensive SO_2 satellite observations available on this eruption, including 3-D information from MIPAS. This is not the case for the Pinatubo eruption, and there are large uncertainties in observations. We will add this explanation on why we chose Nabro.

We also want to clarify, that this study is not a specific study on the impacts of volcanic eruptions. We want to present the new submodel, evaluate it, and provide a default setup to achieve a realistic SO_2 emission plume. We do not concentrate on extreme events, such as the Pinatubo eruption, but we include various eruptions plumes. The goal is to provide a standard setup that includes accurate SO_2 emissions also for studies not focussing on volcanic eruptions. The extreme event of the Pinatubo did not appear to be a suitable candidate for this evaluation.

The introduction could focus more on topics relevant to the rest of the manuscript. E.g. why do you explain ozone chemistry and ash in such detail if it is not relevant to the manuscript? The manuscript provides a very broad discussion about the general impact of volcanic eruptions on climate and atmospheric chemistry, but it does not discuss the problems and current limitations when modeling volcanic eruptions (e.g., spatial & temporal resolution, model agreement/disagreement of past model studies among each other and with observations, sectional versus modal microphysics modules, previous studies which addressed the vertical/horizontal distribution of volcanic emission plumes). Since you submit to "Geoscientific model development" I think a more technical motivation/introduction would be appropriate. I think the introduction should clearly motivate the research so that it is clear to a reader why the submodule was developed and why there is need for the research provided. In my point of view this is not the case.

We thank the reviewer for this suggestion. We chose to focus more on the general volcanic impacts to motivate why this submodel is needed within MESSy, and not necessarily why it is needed in addition to other available volcanic emission routines in various models. We agree with the reviewer, that a more technical focus would improve the introduction, and we will discuss limitations in modeling volcanic eruptions more detailed in a revised version. We did motivate the research by showing, why it is crucial to have this submodel in MESSy, but we can make it clearer, why we did not merely take an existing routine from other models.

Your conclusions just summarize what you have done in your work, including a brief recapitulation of some qualitative results with a brief outlook for further potential applications of the sub-model in the end. However, I think the conclusion section should summarize the key findings of the paper (also some quantitative statements, not only qualitative) and put them into a broader context. The conclusions should demonstrate the importance of the paper and convey the larger implications of the paper to the research field. Additionally, I also find it important to address the limitations of the research/model provided (or more specifically: of the presented sub-module) and highlight aspects identified in your work which require further research and development. I don't think these points are provided in the current version of the conclusion section of the manuscript.

Yes, we mostly summarized our work, but in addition, we also summarize our key findings, e. g. "Horizontal position and emission timing were found to have a minor impact on the long-term SO_2 burden in the stratosphere. Nevertheless, these parameters play a crucial role in detailed process studies during the initial weeks after an eruption. Overall, we conclude that simulations of volcanic eruptions can be effectively performed with the help of 3D-, column, and point emissions. The optimal approach depends on the specific use case, with column emissions excelling in the short-term, and similar performance in the long-term" and " [...] , and we advocate its inclusion in simulations using the MESSy framework focusing on the upper troposphere and stratosphere. For very strong eruptions, it may be beneficial to distribute the emissions over multiple horizontal grid boxes and an extended time period, or adjust the vertical plume width, if discrepancies with observations occur".

We agree, that we can put them into some broader context with previous findings, and potentially point to some limitations and shortcomings, discussed in detail in the Discussion. Additionally, we will outline further necessary research and development in a revised version. However, a technical paper describing a new submodel, evaluating it, and developing a default setup does not need "larger implications of the paper to the research field" but rather vital informations for the people using the submodel. Larger scientific implications to the research field can result from future studies using the submodel.

Similarly, the abstract just summarizes what you did in this study, but you do not provide any results or conclusions or broader implications to the research fields in the abstract, which I think is an essential part of the abstract.

As outlined above, this is a technical paper describing a new submodel, evaluating it, and developing a default setup. The conclusion basically is, that it was developed, it works and it can reproduce observations. There are no real broader implications for the research field, apart from the findings mentioned in the conclusion (see above). However, broader implications are not required in a "model description" manuscript in GMD, as this one is.

An important aspect which is missing in the analysis and discussion of your results is the role of chemical loss of SO_2. In addition to dilution and transport, a (maybe even more) important aspect which determines the SO_2 lifetime is chemical SO_2 loss (i.e., mainly oxidation with OH and O_3). How is this represented in MECCA, and how does this influence your modelled results. You also write that for the Kilauea study, you applied "simplified chemistry". What does this mean? Since chemical loss is very important for the SO_2 lifetime you need to discuss how this affects the results in your study. How is the reaction with O_3 and OH represented in MECCA? Were the O_3 and OH fields also nudged to observations like written in the model description (probably not)? How does this influence the modelled results?

We agree with the reviewer that the chemical loss of SO_2 is important, and actually is the reason for sulfuric acid aerosols to form. In the supplement, we provide the full chemical mechanism for both simulations, the stratospheric one and the "simplified chemistry" for the Kilauea simulation. In a revised version, we will also cover this in the manuscript.

While we treat SO_2 oxidation in both setups as described in the supplement, we do not consider oxidation with O_3. This reaction only takes place in the aqueous phase and is not important for the stratosphere (e. g. Kremser et al., 2016). O_3 and OH are part of the chemical mechanism (see supplement) and are not nudged towards observations. The described nudging affects only the model dynamics using meteorological reanalysis data.

Another aspect which is not discussed is the importance of aerosol microphysics. You show an in-depth analysis of the SO2 plume evolution and then in chapter 4

you suddenly come up with AOD analysis. However, there is an important step missing in between: Aerosol Microphysics. You write that you are using GMXE as an aerosol microphysics module and represent the aerosols using a modal approach. However, you do not provide any information about resulting aerosol burden, aerosol size distribution ect. You only show AOD and extinction (without indicating the wavelengths under consideration). Thus, the whole aerosol microphysics (which is a very important aspects when simulating volcanic emission plumes) is treated as a black box in this manuscript. In my point of view, it is crucial to also show resulting aerosol burden and compare them with observations as well as which other models (e.g. see Brodowsky et al. 2021).

We agree with the reviewer, that aerosol microphysics is a very important aspect for the modelling of volcanic aerosol, and we can provide more detail on the representation in GMXe. Thank you for pointing this out. However, we want to make clear again, that this manuscript is not aiming to evaluate the aerosol microphysics or study the sensitivity of the results to the aerosol microphysics.

We thank the reviewer for pointing out to Brodowsky et al., 2021, and we apologize for not including it in the initial submission. We will provide a comparison of resulting sulfur and sulfate burden in a revised version, and point to the discussed uncertainties. Nevertheless, in our opinion our study has a different focus, as we want to evaluate the EVER submodel. Thus, we look into the uncertainties in the SO_2 emission parameters, while not considering uncertainties in aerosol microphysics in great detail. Our manuscript is a "Development and technical paper" in Geoscientific Model Development, while the Brodowsky et al., 2021 paper is a scientific study in JGR Atmopheres focusing on the evolution of sulfate aerosol, and thus focuses more on the production of H_2SO_4 and the aerosol burden.

We apologize for not mentioning the wavelengths under consideration, and will do so in future versions. Additionally, we will compare our results to previous model results. We decided to focus on comparison to observations of SO_2, as this is mostly influenced by the EVER submodel, and we wanted to show that we can reproduce observed SO_2 mixing ratios. As pointed out, resulting aerosol burden depends on a lot of different factors, the evaluation of which goes beyond the scope of this manuscript. However, we plan to include aerosol burdens as well in a revised version to motivate the evaluation using AOD and extinction.

Another aspect which comes too short in the discussion section is the influence of the spatial and temporal resolution of different processes. It is well known that these aspects are very important for realistic representation of volcanic plumes. While for the troposphere the horizontal resolution is more important in the stratosphere the vertical resolution is more important. For aerosol microphysics the microphysical timestep should be set small enough to realistically simulate nucleation and condensation. You could mention these aspects in the discussion of your results.

Yes, the spatial resolution is very important for the modelling of volcanic eruptions. For that reason, we chose the maximum available horizontal resolution for the tropospheric study of Kilauea (T255), and the maximum available vertical resolution (90 levels) for the stratospheric study as also used as maximum vertical resolution in Brodowsky et al., 2021. The evaluation of different resolutions again goes beyond the scope of the paper, but we can discuss these aspects, yes. Thank you for the suggestion.

We do not distinguish the chemical and microphysical timestep in our simulation, as the general model timestep length is only 8 minutes (we will mention the timestep length in the manuscript), and chemistry and microphysics are calculated in each timestep. This is different to the recent publication by the reviewer,Vattioni et al., 2024, where chemical and microphysical timestep differ. In Vattioni et al., 2024, the reviewer concluded, that in volcanic simulations, first considering condensation and nucleation results in the smallest numerical error. This is the default in GMXe, and thus we did not discuss it. Further reducing the timestep will of course improve the results, but is not computationally feasible. We additionally argue, that the chemical timestep length of 2 hours (in Vattioni et al., 2024) will lead to different errors in microphysics compared to the 8 minute chemical timestep length in our EMAC simulations. Moreover, the main focus of the manuscript is on the initial SO_2 plume, that is not influenced by the microphysical settings.

I also find the manuscript too long. Many aspects which are discussed in the introduction and submodel description are not relevant to the storyline or are not picked up again in the discussion. I suggest shortening substantially and putting part of the text (e.g. description of code and namelists) into the supplement. Also, the structure could be improved. For example, the model description and setup and description of the observations (sect. 3.1 and sect. 3.2) could be a chapter for its own or part of chapter 2. Because the setup described there is also used in chapter 4. The different events simulated here (i.e., Nabro, Kilauea and the 2007-2011 period) seem a little disconnected to each other. Why don't you show a full analysis of only one event (e.g. Nabro), but in more detail including sulfuric acid aerosol burden etc.

Thank you for this suggestion. We agree, that we can shorten the introduction with respect to the discussion of the general impacts of volcanic eruptions. However, we believe that in a technical paper on the implementation of a new submodel, the description of the code and namelist is essential, and is common practice in numerous GMD papers. Moreover, we do not believe that the submodel description in a technical paper has to necessarily follow the storyline of the evaluation, but should instead focus on completeness.

Regarding the restructuring of the manuscript, we agree that we could make 3.1 and 3.2 a separate Section, as the setup is also used in Sect. 4. Nevertheless, we are hesitant to include this in Sect. 2, as we wanted to focus solely on the new submodel in this section.

We understand, that the different events seem to be disconnected. We will motivate the methodology in more detail in a revised version. We chose the different events for the following different reason:

- Single analysis of the Nabro volcano to evaluate the submodel in the stratosphere, and perform sensitivity studies on the emission parameters of the submodel.
- Kilauea study to show the applicability of the submodel for degassing volcanoes
- 2008-2011 period to evaluate the historic namelist setup developed

For this reason, we want to keep the current structure. Nevertheless, we can analyse the Nabro eruption in more detail (including resulting burdens).

References

Brodowsky, C., T. Sukhodolov, A. Feinberg, M. Höpfner, T. Peter, A. Stenke, and E. Rozanov (2021). “Modeling the Sulfate Aerosol Evolution After Recent Moderate Volcanic Activity, 2008–2012”. In: *Journal of Geophysical Research: Atmospheres* 126.23, e2021JD035472. DOI: https://doi.org/10.1029/2021JD035472.

Brühl, C., J. Schallock, K. Klingmüller, C. Robert, C. Bingen, L. Clarisse, A. Heckel, P. North, and L. Rieger (2018). “Stratospheric aerosol radiative forcing simulated by the chemistry climate model EMAC using Aerosol CCI satellite data”. In: *Atmospheric Chemistry and Physics* 18.17, pp. 12845–12857. DOI: 10.5194/acp-18-12845-2018.

Jöckel, P., A. Kerkweg, A. Pozzer, R. Sander, H. Tost, H. Riede, A. Baumgaertner, S. Gromov, and B. Kern (2010). “Development cycle 2 of the Modular Earth Submodel System (MESSy2)”. In: *Geoscientific Model Development* 3.2, pp. 717–752. DOI: 10.5194/gmd-3-717-2010.

Kremser, S., L. W. Thomason, M. von Hobe, M. Hermann, T. Deshler, C. Timmreck, M. Toohey, A. Stenke, J. P. Schwarz, R. Weigel, S. Fueglistaler, F. J. Prata, J.-P. Vernier, H. Schlager, J. E. Barnes, J.-C. Antuña-Marrero, D. Fairlie, M. Palm, E. Mahieu, J. Notholt, M. Rex, C. Bingen, F. Vanhellemont, A. Bourassa, J. M. C. Plane, D. Klocke, S. A. Carn, L. Clarisse, T. Trickl, R. Neely, A. D. James, L. Rieger, J. C. Wilson, and B. Meland (2016). “Stratospheric aerosol—Observations, processes, and impact on climate”. In: *Reviews of Geophysics* 54.2, pp. 278–335. DOI: 10.1002/2015RG000511.

Schallock, J., C. Brühl, C. Bingen, M. Höpfner, L. Rieger, and J. Lelieveld (2023). “Reconstructing volcanic radiative forcing since 1990, using a comprehensive emission inventory and spatially resolved sulfur injections from satellite data in a chemistry-climate model”. In: *Atmospheric Chemistry and Physics* 23.2, pp. 1169–1207. DOI: 10.5194/acp-23-1169-2023.

Vattioni, S., A. Stenke, B. Luo, G. Chiodo, T. Sukhodolov, E. Wunderlin, and T. Peter (2024). “Importance of microphysical settings for climate forcing by stratospheric SO_2 injections as modeled by SOCOL-AERv2”. In: *Geoscientific Model Development* 17.10, pp. 4181–4197. DOI: 10.5194/gmd-17-4181-2024.