

Interactive comment on “Modeling study of the impact of SO₂ volcanic passive emissions on the tropospheric sulfur budget” by Claire Lamotte et al.

Anonymous Referee #1

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This modelling study of the impact of non-eruptive volcanic SO₂ emissions could have been interesting, given the importance of such emissions for the global sulfate aerosol budget. The authors implement a recently developed volcanic emission inventory (Carn et al.) which represents a significant improvement in comparison with the widely-used GEIA inventory. Those inventories are tested using the global model MOCAGE and evaluated against spaceborne SO₂ columns.

Unfortunately, they use GOME-2 SO₂ columns from ACSAF, maybe the worst possible choice of SO₂ satellite data. OMI SO₂ data would have been much more appropriate. The SAF dataset is not even the best GOME-2 dataset. In fact, examination of Figure

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S1 shows two things: 1) the filtering applied to the columns has an disproportionate impact on the columns, and 2) the filtered GOME-2 columns (Figure S1b) have a completely unrealistic distribution. Hot spots are found in every very dry areas on Earth including South Africa, Mongolia, Tibet, Central Australia and Western U.S. This is a strong and obvious artefact. The North China Plain shows a weak enhancement, but much less pronounced than Tibet. This is not credible at all. As far as I know, this dataset has not been validated nor has it been used for any scientific investigation. The correlation coefficient between the model and the data is negative (-0.17) on the global scale, and it is insensitive to choice of the emission inventory. I'm afraid that any conclusion drawn from comparisons with the model are probably useless.

The authors should use a better SO₂ dataset. I do not accept the argument that "only instruments different from those used to set the inventories can be selected for an independent evaluation". On the contrary, it seems imperative to confront the model with OMI SO₂ data and check the overall performance of MOCAGE against those data. It would make the paper much more interesting. Confronting the model with GOME-2 could be interesting as well, but a better dataset would have to be used.

Some additional important comments:

The paper insists several times that "the contribution of volcanic emissions is argued as non-linear on the sulfur species burden". There seems to be quite a confusion regarding the nature of non-linearity. Yes, volcanic SO₂ is longer-lived than SO₂ from other sources, because it is emitted at higher altitudes and is therefore less subject to dry and wet deposition. But this does not make the contribution of volcanic emissions "non-linear". It would be non-linear if the SO₂ emissions would significantly alter their own lifetime (as is the case e.g. for NO_x, due to the strong influence of NO_x on hydroxyl radical concentrations). I don't think this is what the authors mean here. The emphasis on the role of non-linearity should be removed from the paper.

The paper also insists that non-eruptive volcanic emissions were injected at the first

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model level in previous studies. This is not correct. The altitude of the mouth of the volcanoes is of course well known since a long time, and was taken into account already in the early global studies of the sulfur cycle, e.g. Spiro et al. (1992), Pham et al. (1995), Chin et al. (2000). The crater lies generally much higher than the lowest level of the model.

Finally, the article is difficult to read. The text is often unnecessarily long, with many repetitions. The English language could be much improved, see below my suggestions for changes. Many sentences are either illogical, confusing or of little informative value. It is not normal that a reviewer would have to correct so much a scientific paper.

Minor comments

- I. 32 COSPEC: here, make reference to section 3.1 which explains what it is.
 - I. 33 TOMS: make clear that TOMS provided only crude measurements of SO₂ columns.
 - I. 117 "first five levels": indicate the approximate altitude range. Why not injecting emissions at the first level only?
 - I. 142-144 The description of SOA parameterization is very brief, and could be expanded. How well does it perform against organic aerosol observations?
- Table 1 states that the Carn et al. inventory relies on TOMS and OMI, whereas the text mentioned 7 different satellite instruments.
- I. 253-254 "One simulation takes into account only anthropogenic emissions": strange, no biomass burning or natural S emissions? → replace by "The first run (NOVOLC) neglects volcanic emissions." Adapt also the rest of the paragraph.
 - I. 291 "daily mean SO₂": satellites do not provide daily means.
 - I. 295 "thanks for fitting AMF": unclear. As far as I know, the AMF is not fitted.

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Table 3 does not bring much, since the global MNMB is given in the text, and the correlation coefficient is negative. Could be moved to the supplement. The global MNMB is not much interesting given the compensation between very high and very low values apparent in Figure 3.

- I. 360 "We notice small changes in the vicinity of volcanoes where MNMB score is improved": there are many cases where the MNMB is worsened, including Hawaii and islands (Vanuatu?) in the Southern Pacific.
- I. 361 "FGE is better" → "The FGE is slightly improved"
- I. 362 Some comments are needed concerning the negative value of the correlation coefficient.

At this point, I stopped reading, since I think that the GOME-2 dataset (on which this investigation relies) has no scientific value.

Minor (language) comments

- I. 1 Why "Thus"? The sentence remains true even in absence of non-linear behavior.
- I. 3 at the global surface → at the global scale (?)
- I. 4-5 I would rephrase as "the changes induced by the update of the volcanic emissions inventory are studied using the ..."
- I. 7 "degassing" → "degassing emissions"
- I. 8 "uncertainties by volcanoes": what does that mean?
- I. 9 "negligible"
- I. 12 and elsewhere: remove the dot between Tg and yr
- I. 17 "necessity of estimates accurate volcanic sources" → "need for accurate estimates of volcanic sources"

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- l. 20 delete "natural" before "volcanic"
- l. 25 "Plus" → "Moreover"
- l. 30 "to well constrain" : ?
- l. 43 "were not very accurate in quantitative, spatial and temporal detection": weird wording, please rephrase
- l. 44 "used on": ??
- l. 46 Andres and Kasgnoc (1998) work → The study of Andres and Kasgnoc (1998)
- l. 51 "As well": ??
- l. 55 "in its works": ?? "more numerous and qualitative data": ??
- l. 59 "for passive source strength": ??
- l. 61 "high change" → "stark improvement"
- l. 61 "last decades studies" → "studies of the last decades"
- l. 63 "the radiative forcing induced" → "the subsequent radiative forcing"
- l. 67 "on surface species concentration and deposition" → "on the surface concentration and deposition of sulfur species"
- l. 67 "We want": ??
- l. 74 the configuration of simulations with MOCAGE
- l. 76 "updating inventory": ??
- l. 76 "the comparison for"
- l. 77 "Then" → "Next"
- l. 83 "Its use is applied": ??

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- l. 94 "the duration emission" → "the duration of the emission"
- l. 115 "from biomass burning process" → "emitted from biomass burning"
- l. 124 "completed": ??
- l. 158 "It was carried out over a period of about 25 years": I suppose you mean the measurements span 25 years. Please rephrase.
- l. 167 "thanks to the similar molecular structure of SO₂ and ozone": misleading, rephrase or omit.
- l. 167 "Thus": ?? The following sentence is unclear. This could be simplified, as not really necessary.
- l. 178 "constancy" → stability
- l. 178 "Thus" could be omitted
- l. 178-179 "in order to incorporate natural variations due to temporal and even chemical inhomogeneities": confusing. Could be omitted.
- l. 181 "as the one...": replace by "as being among the largest..."
- l. 181 "passive": ??
- l. 181 "For them..." → "For those volcanoes, fluxes (...) supersede the averages"
- l. 185 "Knowing that" → "Since"
- l. 189 "lowest levels" or "lowest level"?
- l. 191 "technological improvements in satellite technology": awkward.
- l. 197 The work of Carn et al. (...) updates and completes the study of Andres and Kasgnoc (1998).
- l. 203 "given is" → "given includes"

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- l. 204 "measured" → "estimated"
- l. 205 "We will..." is somewhat ambiguous. Within this study or later on?
- l. 207 "the daily frequency allows to take into account the eruptions in simulations for the period...": weird statement.
- l. 211 "could distinguish" → "made possible to distinguish"
- l. 222 "every day of the year" → "throughout the year"
- l. 236 "the update of the..." → "the updated..."
- l. 236 "compiles" → "includes"
- l. 237 "spread over the globe": ??
- l. 244 Delete words "lon" and "lat"
- l. 246 "volcanic one" → "volcanic source"
- l. 246 "The same global annual sulfur emissions are computed for all other sources": of course since the same inventories are used!
- l. 248 "emissions are" → "emissions amount to". You don't need two significant digits after the decimal point, one is enough.
- Table 1 legend "Summary information on" → "Summary of"
- Table 1 "Nb of volcano" → "Number of volcanoes"
- l. 253 "characteristics" → "main features"
- l. 256 "However, one injects the volcanic SO₂ emissions" → "In simulation CARN, volcanic emissions are injected". Adapt also the rest of the sentence
- l. 261 "in altitude" → "in the vertical"
- l. 262 "Then": ?? The entire sentence is weird. You could drop it since you explain

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what you do in the following sentences.

- l. 263 "The CARNALTI run is expected to provide the best..."
- Figure 1 legend: drop "annual" (since monthly values are shown). "anthropogenic" or "other emissions"?
- l. 269 lowest eruptive emission flux (Carn et al., 2016)
- l. 269 "is negligible". That sentence could be dropped.
- l. 270 Why the upper-case AND?
- l. 270 This sentence is weird, not really useful.
- l. 273 "adds": ??
- l. 275 "referenced": ??
- l. 276 "counts": ??; "into": ??
- l. 277 "are" → "amount to". Use only one significant digit for the totals.
- l. 279 "current": the use of this word for the previously used inventory is weird. Replace maybe (here and elsewhere) by "previous"
- l. 280 "inventory against" → "not accounted for by". Delete "one".
- Figure 2 Legend "rounds" → "circles"
- l. 284-285 Weird sentence, provide more direct formulation.
- l. 285 "benefit". The sentence is true but too obvious.
- l. 289 "indirectly correlated to SO₂": ??
- l. 299 "Plus" → "In addition"; "presence of offsets" → "offsets" (?); "lead" → "leads"; "criteria" is plural, replace by "criterion" (if meant as singular)

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- l. 301 "subtracted at" → "subtracted from"
- l. 311 "low confident" → "low-confidence"
- l. 311 "filtered" → "filtered out"
- l. 318 "we can use several statistical metrics": delete, and merge with next sentence "we use the fractional bias,..."
- l. 335-337 This paragraph could be omitted. Delete "Therefore" from the next paragraph.
- l. 340 "Plus" → "Furthermore". But I don't understand well the rest of the sentence. Rephrase.
- l. 343 You might drop the word "inventory" after the reference. Same remark applies elsewhere in the text.
- l. 342 Drop "The" before Zone 1. Same elsewhere.
- l. 346 "are" → "amount to"
- l. 354 "counting"??
- l. 358 "higher" → "less negative"
- l. 359 "againts" → "against"
- Table 4 "Coorelation" → "Correlation"; "specifics" → "specific"
- l. 698: the link does not work

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