Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2017-538-RC2, 2017

Atmospheric Chemistry and Physics Discussions

Interactive comment on "MIPAS observations of volcanic sulphate aerosol and sulphur dioxide in the stratosphere" *by* Annika Günther et al.

Anonymous Referee #2

Received and published: 11 August 2017

This study presents new measurements of aerosol volume densities and H2SO4 concentrations for 2005 to 2012 as obtained from MIPAS on-board ENVISAT. Using a chemical transport model (CTM), they also investigate the evolution of volcanic SO2 emitted from two volcanic eruptions in the northern mid-latitudes, eruptions of Kasatochi and Sarychev. This is a good paper that complements existing aerosol measurements and existing studies on investigating the volcanic eruption of Kasatochi and Sarychev. The paper presents new data sets that will be of interest to the readership of ACP. The paper would benefit from greater clarity in writing and from providing more information on the CTM model simulations, on the methodology of obtaining SO2 mass that has been used in the CTM simulations and on the bias correction that has been applied to the MIPAS data. After addressing my comments stated below, I recommend

Printer-friendly version

Discussion paper

the paper to be published in ACP.

General

The paper contains a number of spelling and grammatical errors. I only point out a few of them below and I would encourage the authors to re-work through the paper and correct all the errors. I would like to point out that the spelling of sulfur throughout the paper is incorrect. The journal guidelines clearly state: “In accordance with IUPAC, it is our house standard to use the -f- spelling for sulfur (instead of sulphur) and related words for all varieties of English.”

I noticed that the authors use abbreviation/acronyms without defining them throughout the paper. I would encourage the authors to have a careful look through the paper and provide the definitions for the abbreviations used e.g. H2SO4 in the abstract and introduction. As stated in the journal guidelines, abbreviations “. . . need to be defined in the abstract and then again at the first instance in the rest of the text”.

I know that it seems commonly accepted to write ‘data is’ but data is the plural of datum and therefore it should read ‘data are’. Please check the wording throughout the paper. Also, ’dataset’ should be corrected throughout the paper to ’data set’.

The abstract would benefit from a clearer structure. The authors describe the measurements briefly, go into the case study and then back again to the measurement. A clear ‘story line’ of what they did and what they have found is missing or it is not clear when the authors refer to the case study and then to measurements.

Specific comments

Page 1, ln 13: “. . . on board of the Environmental Satellite”

Remove ‘of’ and include (Envisat) at the end as I believe this satellite is mostly known by its acronym.

Page 1, ln 14/15: “The MIPAS aerosol dataset has been corrected for a possible

altitude-dependent bias by comparison with balloon-borne in situ aerosol measurements at Laramie, Wyoming."

I'm not sure about the word 'possible' in this sentence. Is there a bias or not and if there is why 'possible'? It is not clear from reading this sentence how the bias was corrected and what the Laramie measurement have to do with it? Was the bias discovered when comparing the measurements to the balloon measurements or was the comparison used to correct the bias, or both? This sentence needs to be made clearer. Please change the wording of 'possible altitude-dependent bias' throughout the paper.

Page 1, ln 15/16: 'The MIPAS data of stratospheric sulphate aerosol is linked to MIPAS observations of sulphur dioxide (SO2) with the help of Chemical Transport Model simulations.'

Replace 'is' with 'are'. Also, what do you mean by saying 'data are linked to MIPAS SO2 observations'? How can you link observations with CTM simulations?

Page 1, ln 16/17: 'We investigate the production of sulphate aerosol. . .'. '. . .and its fate from volcanically emitted SO2 for two volcanic case studies:'

Production of sulfate aerosol in the stratosphere I assume? For this you are using the CTM? To use 'its fate' is this sentence seems to be a rather odd word choice. Could the authors replace 'fate' throughout the paper?

Page 1, ln 20: 'While sedimentation of the sulphate aerosol plays a role, we find that the dominant mechanism controlling the stratospheric lifetime of sulphur after these volcanic eruptions at mid-latitudes is transport in the Brewer-Dobson circulation.'

This sentence needs to be reworded. How about: '. . . the lifetime of stratospheric sulfur is controlled mainly by the Brewer-Dobson circulation'.

Page 2, ln 5/6: 'Hofmann et al. (2009) observed an increase of stratospheric aerosol and speculated that this is due to anthropogenic emissions.'

Change ‘increase of’ to ‘increase in’. Please also include ‘increase in stratospheric aerosol load. . .’ (or abundances). Use ‘suggested’ rather than ‘speculated’.

Page 2, ln 6/7: ’Newer studies, however, show this increase to be connected more likely to a series of smaller and medium sized tropical volcanic eruptions (e.g. Neely et al., 2013).’

Reword to: ’. . . show that this increase is likely to be connected to a number of small and medium sized volcanic eruptions located in the tropics.’

’Following Vernier et al. (2011), the increase of stratospheric aerosol levels since 2002 is connected to a series of moderate eruptions of volcanoes especially in the tropics.’

This seems repetitive and should be combined with the sentence before.

Page 2, ln 9: ’These volcanoes directly injected sulphur up to 20 km into the stratosphere’

Not clear what ‘these’ refers to and here a reference is needed that states that sulfur got injected into the stratosphere.

Page 2, ln 10: remove ’,’ after (2014).

’. . .noticed a strong contribution of aerosols in the lowermost stratosphere of the mid- and high latitudes to the volcanic aerosol forcing during the last decade’

It is not clear to me from this sentence what the authors are trying to say and what the message is.

Delete sentence: ’Understanding of stratospheric sulphur, its sources and sinks, and the processes involved in its conversion and transport is important in the framework of proposed climate engineering schemes (e.g. Niemeier and Timmreck, 2015; Rasch et al., 2008).’

Not sure why this is mentioned here and how that relates to this study.

Page 2, ln 14: The authors should define ‘background conditions’, i.e. that they mean ‘non-volcanic’ conditions.

Page 2 ln : Remove ‘E.g.’

’Chin and Davis (1995), Thomason and Peter (2006), Bruhl et al. (2012),and Sheng et al. (2015), agree on a major contribution of OCS.’

Contribution to what?

’However, its exact contribution to stratospheric aerosol during background conditions is still in discussion.’

Replace with ’. . . the magnitude to which OCS contributes to stratospheric aerosol loading . . .’

’During volcanically perturbed times volcanically emitted SO2 is the dominant source for stratospheric sulphate aerosol and causes most of the variability in the stratospheric sulphur content.’

The word ‘volcanically’ is used twice, why not just saying: ”Volcanic eruptions are the dominant source for . . .”. Replace ‘content’ with ‘loading’. Are the authors referring here to the stratospheric sulfur concentrations or aerosol loading?

’In volcanic emissions, SO2 is the third most abundant emitted gas, after water vapour and carbon dioxide (von Glasow et al., 2009).’

Why is this relevant?

Page 2, ln 23: ‘. . . is useful’. Useful for what?

’From MIPAS several datasets that are relevant to the stratospheric sulphur content are already available.’

Replace ‘content’ with ‘concentrations’ or ‘loading’. Replace ‘datasets’ with ‘data sets’ and please correct throughout the paper. Can the authors please clarify why MIPAS

measurements are relevant to the stratospheric sulfur loading? I believe the word choice here is misleading. Do the authors mean that MIPAS measurements are important to estimate the stratospheric sulfur loading?

Page 2: 'Here we present an additional dataset of sulphate aerosol from MIPAS, and combine the MIPAS SO2 and liquid-phase H2SO4 measurements with Chemical Transport Model (CTM) simulations to analyse the consistency of the two datasets, and the fate of volcanically emitted sulphur.'

Again, 'fate' is an odd word choice. It is not clear from this sentence what the CTM was used for and why. Are the authors here talking about two or three data sets?

Delete 'This paper has several purposes.'

Page 3, ln 3: Why did the authors choose the 2005-2012 period?

Page 3: 'We analyse MIPAS observations of SO2 and stratospheric sulphate aerosol in comparison to CTM simulations, and study the sulphur mass contained in SO2 and sulphate aerosol, together with the transport of their volcanic plumes.'

This sentence needs to be re-worded. It is not clear that this analysis is related to the volcanic eruptions stated in the sentence before.

'Finally, in Sect. 5 we draw final conclusions on the consistency between the MIPAS SO2 and the new MIPAS sulphate aerosol dataset,'

Does this only relate to the data sets during the volcanic eruptions? Either replace 'Finally' or 'final'.

Page 3, ln 23: It would be good if the authors could state the time period of the SO2 data set they are using in this study so that the reader doesn't need to look into Hoepfner et al. (2015).

Page 3, ln 28: Replace 'bias of' with 'bias between'

Page 3, ln 30: Replace ‘within’ with ‘between’

’. . .when due to aerosol-related sampling artefacts the total mass of SO2 was found to be strongly underestimated (Hopfner et al., 2015).’

Re-word to: ’ when the total mass of SO2 was found to be strongly underestimated due to aerosol-related sampling artefacts (..).’

’Their study comprises a dataset of volcanically emitted SO2 for 30 volcanic eruptions, as seen in the MIPAS measurements’

Not clear what ‘their’ refers to (I assume that is the study by Hoepfner et al.). Suggest to reword this sentence and also to write ‘as observed by MIPAS’ rather than ‘seen in the MIPAS measurements’.

Should Section 3 be moved forward to 2.1.3? The structure of this paper is not clear to me. Section 2 was labelled ‘Data sets and Methods’ but then Section 3 is ‘MIPAS aerosol data set’. That is confusing.

Page 4: ’The in situ measurements were completed with balloon-borne University of Wyoming optical aerosol counters and consist of size resolved aerosol concentrations from the surface to approximately 30 km.’

Replace ‘completed’ with ‘made’. . . Suggest to change sentence to: “Size resolved aerosol concentration measurements from the surface to approx. 30 km altitude were made with . . .”

Page 4: Reword sentence ’The latest style of the three was used initially in 2006, became the standard Laramie instrument in 2008, and was flown on quasi-Lagrangian balloons in Antarctica in 2010 (Ward et al., 2014).’

Something is missing in this sentence.

Page 4, ln 12/13: ’For the MIPAS validation the Wyoming measurements were confined to those made with this final instrument (Laser based counter, LPC), which measures

particles with radii > 0.08–4.2 μm in eight size classes."

Why were the data from this LPC used? Why not any measurements from other OPC measurements before 2006?

Page 4, ln 15 and following: 'To derive geophysical quantities from the size resolved aerosol concentration measurements it requires fitting a size distribution to the data. In the past this has been done by choosing a subset of the measurements to fit either a unimodal or bimodal lognormal size distribution. The final size distribution selected is from that subset of the measurements which minimizes the root mean square error when the fitted distribution is compared to all the measurements. This approach has recently been changed to use laboratory measurements of the counting efficiency at each channel and then search the lognormal parameter space for the lognormal coefficients, which minimizes the error of the fitted distribution compared to the measurements.'

How does the 'new' approach compare to the 'old' approach? How different are the measurements when derived with the new method compared to the old approach? And why is the new approach not used but mentioned here?

Page 4, ln 24: 'The isentropic Chemical Transport Model used in our study. . .'

Delete 'isentropic'. It is a CTM using isentropic levels as vertical coordinates but does that mean it could not be used differently? Include CTM acronym.

Page 4, ln 30: What are the initial OCS concentrations and where were they taken from? It would help the reader if the chemical reactions included in the CTM would be listed somewhere. Does the model include liquid and gas-phase chemistry? Are you only considering chemistry in the stratosphere? You state 10 to 55 km, and depending on where you are on the globe, 10 km could still be in the upper troposphere. So does cloud uptake of SO2 play a role here? If not, why not?

Page 5, ln 20: Do the authors believe that it will be clear to the reader what MI-

PAS/Balloon is in this context, especially since it is not mentioned before? I would suggest to add more information on this here and also to refer to it at MIPAS-B.

Page 6, ln 2: Remove 'E.g.'

Figure 3: Is it possible to show the uncertainties on the in situ measurements? What does LCP 2m, 1p and 3m stand for? The standard deviations on the mean values are shown but that is not the uncertainty on the measurement which would be interesting to have a look at? Wouldn't it be more appropriate to calculate the uncertainty on the mean using the measurement uncertainties on each datum that went into the mean calculation?

Page 10, ln 3: 'In Fig. 4a the standard errors of the mean show the uncertainty of the bias.'

I don't understand this statement. How does a standard error of the mean profile relates to the uncertainty of the bias? This needs to be clarified.

Figure 3 shows a strong signal at around 18 km in the balloon measurements on 28.07.2011 which does not seem to reflect in any way in the mean and its standard error shown in Figure 4a. Is that expected? This takes me back to the point that the mean values and their standard error do not take into account the uncertainty on each measurement.

What did the authors do to de-bias MIPAS measurements shown in Figure 3 and 4? There are some words around it on Page 10 (ln 5 to 10) but this didn't answer the question about what the authors actually did.

Page 13, ln 1: 'Sulphur is released from OCS mainly in the tropics at altitudes between about 25 and 35 km (Bruhl et al., 2012) and the sulphate aerosol that is built is transported towards mid-latitudes and lower altitudes.'

Is that statement still true with new publications (e.g. Lennartz et al. (2017)) about OCS being published?

Page 13: 'In the mid-latitudes of the Northern Hemisphere, the sulphate aerosol is increased during boreal summer at around 10–12 km.'

I'm not sure if that is a strong enough signal as the Northern Hemisphere is strongly disturbed by volcanic eruption according to Figure 5. What could be a cause for this increase?

Figure 5: There is a strong signal at altitudes 18 to 22 km (around 2007), i.e. enhanced H2SO4 concentrations which cannot be seen in the SO2 concentrations. Do the authors have an explanation for this signal in the tropics that high up?

Page 14, ln 8: Is 'sedimentation radius' the right term to be used? I don't think it is (I think the authors mean the radius of the aerosol) and the authors might want to think about rewording this sentence and where appropriate throughout the paper.

Page 14, ln 9: 'Good accordance between the modelled and measured SO2 masses is essential to test, . . .'

This sentence is rather odd and 'accordance' should be replaced with 'agreement'?

Page 14, ln 11: 'In Table 1, SO2 masses for three altitude regions. . .' Include 'injected SO2 amounts for three altitude regions as used in the CTM simulations. . .' The authors do not explain why and how they have chosen these amounts. Please clarify.

Page 14, ln 12: 'The simulations result in good agreement between measured and modelled SO2.'

Can the authors please clarify what simulations they are talking about? What is the simulation period? What the time step? I feel that there are more information required about the CTM simulations.

Page 14, ln 14: 'This method was applied as in the first month after the eruption MIPAS underestimates the SO2 (Hopfner et al., 2015).'

I don't understand what the authors are trying to say here. This method was applied

in their study or the study of Hoepfner et al., (2015). Why is it important to know how Hoepfner et al. derived the mass? How did the authors derive the SO2 mass used for their study? Could the authors please clarify why they didn't use the same values as Hoepfner et al. and how they derived their values for SO2 mass injected by the two volcanoes? And why do they not provide any uncertainties on their values?

Figure 6: Wouldn't a sedimentation radius of 0 mean that there are no aerosols? Do the authors mean that aerosol doesn't get lost through sedimentation? I don't understand why the eruption of Redoubt (23 Mar 2009, 60.5° N/152.7° W) is included here but was never mentioned before? Also, it is not clear if additional SO2 has been injected in the model due to this eruption (I suspect not). If the authors want to include Redoubt in the figure because there is a signal in the measurements, they should state that clearly in the text.

What in the CTM causes SO2 to be lost so much faster between 10.5 and 14.5 km compared to 14.5 and 18.5 km? Again, I think more information about the CTM is required. Can the authors explain why for a radius of 1μm the H2SO4 loss is leveling off earlier (around November) between 18.5 and 22.5 km than compared to the other altitude ranges? What is causing the second peak (around Dec) in H2SO4 between 10.5 and 14.5 km? This peak is also seen in the MIPAS measurement but shifted by half a month. The peak in SO2 as modelled by the CTM is shifted compared to the peak in MIPAS SO2. Can the authors explain why that is?

Page 18, ln 15: '. . .due to its reaction with OH, and sulphate aerosol is consequently built.'

This sentence needs to be reworded – use formed rather than build.

Page 18, ln 23/24: '. . .the modelled sulphur mass without sedimentation already compares rather well with the measured sulphur mass.'

Looking at Figure 6d to f, are the authors really saying that the no-sedimentation run

Printer-friendly version

Discussion paper

compares well to the measurements, especially in 18.5 to 22.5 km region? I do not agree with this statement and their conclusions drawn from this. The fit to the measurements is better for the runs where sedimentation was considered.

Figure 7: Again why is the Redoubt eruption included in this Figure. This is the first time where the authors mention the simulation periods. This has to come earlier and it has to be described in the text. Why is the pattern for H2SO4 (CTM simulation) for the Kasatochi eruption so different from the Sarychev eruption? For the Kasatochi eruption the H2SO4 concentrations are lower than for the Sarychev eruptions although similar amounts of SO2 where injected? How much of the simulated Kasatochi 'double-plume' is made due to the choice of the SO2 mass being injected at different altitude levels? The injection heights of SO2 for the Sarychev eruption seem not appropriate when compared with the measurements. The model simulations show high SO2 concentrations from 10 to 19 km while in the observations the range goes from 11 to 16 km. Did the authors conduct any sensitivity studies regarding the injection height and the impact on their results?

Page 19: "and reaches 10 km after a few months."

The upper plume doesn't reach 10 km after a few months as shown in Figure 7. It is moving down but by doing so that sulfur concentration reduces. Or are the authors talking about the model simulations?

Figure 8: The CTM model shows about 1 month lag before SO2/H2SO4 is seen in the tropics compared to the observations which show a signal right from the beginning although the signal in the observations might not be from the volcanoes investigated here? The signal in the measurements in August must be from a different volcano? For the Sarychev eruption, the model seems to show the signal in the tropics earlier than it is seen in the measurements. Why could that be? The lag (time lag for when the eruptions is seen in the tropics) seems to be reduced for the Sarychev eruption? The model seems to overestimate the peak amount of SO2+H2SO4 compared to the

Printer-friendly version

Discussion paper

observations. Any explanation for that given that the model agrees with the peak values of measurements better between 30 and 90N?

Figure 9: For the CTM simulations, it looks like SO2 is immediately enhanced at lower latitudes (down to 15deg N) at the time of the eruption, especially for the Sarychev eruption. Why is that? SO2 is much more confined to the NH in the observations. How is the SO2 emitted by the volcanoes injected into the model? The authors state that 'SO2 is uniformly distributed to the grid boxes per altitude range' but then I don't understand why the picture for Sarychev at 12 and 14 km is so different from the picture for Kasatochi given that SO2 was uniformly distributed between 10 and 14 km (just looking at the time of the eruption, realizing that things will change after some time)? I find the differences in the latitudinal distribution of SO2 between the observations and the model interesting and would like the authors to comment on that.

Page 22, ln20: 'Especially at low altitudes we find a mixing barrier at $\sim$30° N, with a strong gradient between low values in the tropics and high values in the extra-tropics, which weakens towards higher altitudes.'

Which seems to be more pronounced in the measurements than in the CTM, especially for SO2. Do the authors have any explanation for the 'leak' towards the equator in the model, especially in July/Aug after the Sarychev eruption?

Page 22, ln 25: 'An additional transport process starts at an altitude of about 18 km in the case of Kasatochi and $\sim$16 km in the case of Sarychev (Fig. 9 and 10).'

What 'additional' transport process are the authors referring to here?

Figure 10: Enhanced H2SO4 concentrations reach further South in the model simulations than the observations show (at 18 km). In the observations H2SO4 is more confined to latitudes between 15 and 60deg N while for the model simulations, enhanced H2SO4 concentrations reach the equator. Why the difference?

Page 23, ln 12: 'The weaker southward transport in the case of the Kasatochi eruption

Printer-friendly version

Discussion paper

that starts at higher altitudes, compared to the Sarychev eruption, could be due to the eruption having been later during the monsoon season, leading to enhanced southward transport by the Asian summer monsoon for a shorter period of time.'

This sentence is confusing... 'weaker transport leading to enhanced transport'? Could the authors please clarify this sentence. I think I know what the authors mean but they need to clarify which eruption they are talking about in the second part of the sentence 'could be due to the eruption'.

Page 23, ln 20: 'In this study a new dataset of MIPAS/Envisat global aerosol volume densities and liquid-phase H2SO4 VMR distributions is presented for 2005 to 2012. . .'

Are the authors not talking about two data sets that are presented? Replace 'is' with are and use data sets instead of dataset.

Page 24: 'The new H2SO4 aerosol observations enable us to further constrain the total sulphur emitted into the stratosphere by the Kasatochi and Sarychev eruptions and to revise our previous estimates that were based on SO2 observations only.'

So what is the new estimate of total sulfur emitted into the stratosphere by both volcanoes? It would make sense to include that in the conclusions.

Page 24, ln 6: '. . .under OH background conditions'.

What do the authors refer to when saying 'OH background conditions'? That is not clear to me.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-538, 2017.