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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.

Annika Günther et al.

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We thank the reviewer Hugh Pumphrey for his helpful and constructive comments which we address in detail below.

The notation is as follows: P5L27 means page 5, line 27.

General remarks

- The subject is an advance in knowledge, appropriate to the journal, and should be accepted, subject to minor corrections.

- The written English is clear and unambiguous, but has a rather stilted style and a sprinkling of grammatical errors. I note a couple of these below, but this is not a full proof-read.

  Thank you for these corrections. We will try our best to improve the manuscript in this respect.

- The figures are generally clear and well made; I have only a few suggestions for corrections.

  We will take all those into consideration for the final version.

Specific corrections

- All pages: It grieves me to point it out, as, to me, the -f- spelling of "sulfur" is a horrid Americanism which grates on the eye. But the journal's English guidelines state that . . . it is our house standard to use the -f- spelling for sulfur (instead of sulphur) and related words for all varieties of English.

  Thank you very much for this reminder, we will use the "sulfur"-spelling in the revised version of the paper.

- P3L16: The authors note that they use only the second of the two measurement periods, but do not spell out why. Was it not possible to estimate SO_2 from the first period data? Were there no volcanoes of interest during that period?

  The SO_2 dataset by Höpfner et al. (2015) comprises retrieved sulphur dioxide profiles for both measurement periods. However, the first period is not considered within this study. We aimed at investigating two of the major mid-latitudinal eruptions (Kasatochi in 2008, and Sarychev in 2009) during the MIPAS measurement period from Jun 2002–Apr 2012. Volcanic eruptions during the first period only injected SO_2 masses of below 100 Tg to 10–22 km (Höpfner et al., 2015). Furthermore, the much longer second measurement period (Jan 2005–Apr 2012) is characterised by a better vertical and horizontal resolution due to the denser vertical and horizontal limb sampling. Future work will be invested into the retrieval from the first period (Jun 2002–Mar 2004) in order to get an aerosol dataset covering the whole MIPAS lifetime.

The revised version of the paper will include the following sentences on P3L16: “Here we concentrate on the data from the second and longer measurement period (Jan 2005-Apr 2015), as the major mid-latitudinal volcanic eruptions between 2002-2012 occurred during this period. Furthermore, this measurement period is characterised by an improved vertical resolution, especially in the altitude region of the upper troposphere and lower stratosphere.”

- P5L27 “The sulphur . . . builds H_2SO_4” The wording of this sentence and the use of the word “builds” in particular seems rather odd. A possible alternative wording is “The sulfur released from volcanic SO_2 reacts with OH to form H_2SO_4.”

  This will be changed in the revised version.

- P7, Figure 1: The vertical axis of the graph is not labelled and it is not clear to me whether it applies both to the refractive index curves and to the transmission curve.

  Thank you for making us aware of the missing label. The labels will be included in the revised version.

- P9L16–18: I would remove the comma after “Both” and insert one after “increasing temperatures”.

  This will be changed in the revised version.

- Figure 3: The caption does not explain the difference between LPC 2m, LPC 1p and LPC 3m.

  In the caption of Fig. 3, we will clarify that the colour coding for the LPCs means that different Laser Particle Counters have been used for the measurements. “... measured by Laser Particle Counters (LPCs). Different LPCs have been used (colour-coded).”

- Figure 5: It would be preferable to repeat the table of volcano names somewhere in this paper, rather than referring the reader to a different paper. Also, the levels in the filled contour plot are the rather odd choice of 100/7 units. The colour scale itself is a better choice than the dreadful “jet” scale that too many people still use. But I feel that there might nevertheless be a better choice. In particular, I feel that it would be better for the colours at the upper end to become paler (e.g. red → magenta → almost-white) rather than tending towards a purple colour which is very close to the blue at the bottom of the scale. In making any such change it should be ensured that adjacent colours are clearly distinguishable from each other. (This is currently the case except, perhaps, for the shades of blue around 200 ppbv.)

  A list of abbreviations is going to be added to the caption. Furthermore, the purple colours will be removed from the colour-scale in all contour plots, and we will consider updating the levels of the contour plots to match better to the values shown in the colour-bars.

- P13L2: “built” is rather an odd word choice. Maybe “produced” would be better.

  This will be changed in the revised version.

- P15, table 1: Pumphrey’s two estimates for Sarychev are the wrong way round, and one of them is missing its error. It should be 571±42 above 147 hPa, and 1160±180 above 215 hPa.

  Thank you very much for this remark, will be updated in the revised version.

- P22L19: Remove comma after “Both”.

This will be changed in the revised version.

- P24L11: “were” should be “where”.

  This will be changed in the revised version.

- P24L19: “hereby” should perhaps be “thereby”

  This will be changed in the revised version.

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

Atmos. Chem. Phys. Discuss.,
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Atmospheric Chemistry and Physics
Open Access
Discussions

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

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We thank Referee #2 for the detailed and constructive comments (in black). We address the suggested improvements in detail below (in blue).

This study presents new measurements of aerosol volume densities and H_2SO_4 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 SO_2 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 SO_2 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 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.

  Thank you for the error–corrections given in the following. We will take all those into consideration for the revised version, and will try our best to improve the manuscript in this respect.

  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."

  Thank you very much for this reminder, we will use the "sulfur"-spelling in the revised version of the paper.

- 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. H_2SO_4 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".

The updated version of the manuscript will contain the definitions for abbreviations, both in the abstract and main 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'.

  This will be corrected throughout the paper.

- 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.

  Thank you for this advice.

  We will change the structure of the abstract as follows: on page 1, ln 15–16 ("The MIPAS data of stratospheric sulfate aerosol are linked to MIPAS observations of sulfur dioxide (SO_2 with the help of Chemical Transport Model (CTM) simulations.") will be deleted. Instead, on page 1, ln 19 "... during boreal summer. 'With the help of Chemical Transport Model (CTM) simulations for the two volcanic eruptions, we' show that ..." will be added.

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.

  This will be changed in the revised version.

- 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 measurements 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.

  MIPAS aerosol data show a bias in comparison to the in situ data. The de-biasing is made based on this comparison. The word 'possible' will be deleted in the revised version (throughout the paper).

  The information on page 1, ln 14–15 will be changed to: "In comparison to balloon-borne in situ measurements of aerosol at Laramie, Wyoming, the MIPAS aerosol data have a positive bias that has been corrected, based on the difference to the in situ data." On page 23, ln 23 we will replace "The MIPAS aerosol volume densities have been corrected for possible instrumental radiance baseline offsets by comparison to coincident balloon-borne in situ observations from Laramie, Wyoming." by "The MIPAS aerosol volume densities have been corrected for a positive bias in comparison to coincident balloon-borne in situ observations from Laramie, Wyoming. This bias is supposed to be caused by instrumental radiance baseline offsets."

- Page 1, ln 15–16: 'The MIPAS data of stratospheric sulphate aerosol is linked to MIPAS observations of sulphur dioxide (SO_2) with the help of Chemical Transport Model simulations.'

  Replace 'is' with 'are'. Also, what do you mean by saying 'data are linked to

MIPAS SO_2 observations'? How can you link observations with CTM simulations?

'is' will be replaced by 'are'.

On page 1, ln 15–16 “The MIPAS data of stratospheric sulfate aerosol are linked to MIPAS observations ... simulations.” will be deleted. Instead, “'With the help of Chemical Transport Model (CTM) simulations of the two volcanic eruptions, we' show that the MIPAS sulfate aerosol and SO_2 ... .” will be added on page 1, ln 19.

- Page 1, ln 16–17: 'We investigate the production of sulphate aerosol ...'. '... and its fate from volcanically emitted SO_2 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' in this sentence seems to be a rather odd word choice. Could the authors replace 'fate' throughout the paper?

  We study the sulfate aerosol predominantly in the stratosphere, right, and the CTM and MIPAS data are used in the case study on the two volcanic eruptions to do so.

  For clarification we will change the manuscript as follows: “... 'stratospheric' sulfate aerosol ...” will be added to the sentence. Page 1, ln 15–16 “The MIPAS data of stratospheric sulfate aerosol are linked to MIPAS observations ... simulations.” will be deleted. Instead, “'With the help of Chemical Transport Model (CTM) simulations of the two volcanic eruptions, we' show that the MIPAS sulfate aerosol and SO_2 ... .” will be added on page 1, ln 19.

  We will replace the word 'fate' throughout the paper. In this special case no replacement will be done, but “and its fate” will be deleted. (“We investigate the production of sulfate aerosol from volcanically emitted SO_2 ...”) Depending on the sentence, 'fate' is replaced by 'evolution' or 'development'.

- 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 mainly controlled by the Brewer-Dobson circulation'.

  This will be changed in the revised version.

  (“While sedimentation of the sulfate aerosol plays a role, we find that the long-term decay of stratospheric sulfur after these volcanic eruptions at mid-latitudes is controlled mainly by transport in 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'.

  This will be changed in the revised version.

  (“Hofmann et al. (2009) observed an increase in stratospheric aerosol load and suggested that this is due to anthropogenic emissions.”)

- 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.

This will be changed to: “Newer studies, however, show that this increase is likely to be connected to a number of small and medium sized volcanic eruptions especially in the tropics (e.g. Neely et al., 2013; Vernier et al., 2011)”

- 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.

  This will be changed to: “During the last decade several volcanoes directly injected sulfur up to 20 km into the stratosphere (Vernier et al., 2011).”

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

  The comma will be removed in the revised version.

- ’... 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.

  To clarify what we intended to say the sentence will be replaced by: “Ridley et al. (2014) and Andersson et al. (2015) emphasise the importance of volcanic aerosol in the lowermost stratosphere at mid- and high-latitudes on the total volcanic aerosol forcing during the last decade. Their studies show that stratospheric altitudes below ∼15 km (380 K isentrope), which are not represented in many of the aerosol data sets, need to be taken into consideration when studying the global radiative forcing generated by volcanic eruptions in the extra-tropics.”

- 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.

  This information might be misplaced here and will therefore be deleted.

  Instead on page 1, ln 2 we will add the following: “... for climate change modelling studies. ’Increased interest in stratospheric sulfate aerosol is also connected to its potential use in climate engineering schemes (e.g. Niemeier and Timmreck, 2015; Rasch et al., 2008).’ ” as we think that a reference to climate engineering studies should be contained in this paper.

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

  In the revised version we will add this information “background / non-volcanic conditions”.

- Page 2 ln : Remove ’E.g.’

  This will be removed in the revised version.

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

  Contribution to what?

  We refer to the contribution of OCS to stratospheric sulfate aerosol.

  “contribution of OCS ’to stratospheric sulfate aerosol’.” will be added in the revised version.

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

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

This will be changed in the revised version.

- 'During volcanically perturbed times volcanically emitted SO_2 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 say: "Volcanic eruptions are the dominant source for ...". Replace 'content' with 'loading'. Are the authors referring here to the stratospheric sulfur concentrations or aerosol loading?

  We are referring to both, the stratospheric sulfur and aerosol loading.

  The sentence will be changed to: "By emitting SO_2, volcanic eruptions are the dominant source for stratospheric SO_2 (direct) and sufate aerosol (indirect) under non-background conditions, and cause most of the variability in the stratospheric sulfur loading."

- 'In volcanic emissions, SO_2 is the third most abundant emitted gas, after water vapour and carbon dioxide (von Glasow et al., 2009).'

  Why is this relevant?

  This additional information on volcanic emissions will be deleted in the revised version.

- Page 2, ln 23: '... is useful'. Useful for what?

  A combination of observations and model simulations is useful for studies of stratospheric sulfur. However, we decided on deleting this sentence in the revised version.

- '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.

  This will be changed in the revised version.

  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?

  You are right, the word choice is misleading.

  "From MIPAS several data sets 'of trace gas species' that are relevant to 'study' the stratospheric sulfur loading are already available." will be added.

- Page 2: 'Here we present an additional dataset of sulphate aerosol from MIPAS, and combine the MIPAS SO_2 and liquid-phase H_2SO_4 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?

  Basically we are talking about two data sets here. The MIPAS SO_2 and H_2SO_4 data sets. MIPAS SO_2 is retrieved as volume mixing ratios and the aerosol data set consists of aerosol volume densities (also converted into VMR). These are compared to each other (2005–2012), and in a case study on the volcanic eruptions of Kasatochi and Sarychev CTM simulations are included, to analyse whether the measured MIPAS SO_2 after the eruptions can lead to the enhancements as seen in the MIPAS H_2SO_4 (qualitatively and quantitatively), and to study the transport patterns of the volcanic plumes.

  The sentence will be replaced by the following: "Here, we present a new data set of sulfate aerosol volume densities (AVDs) retrieved from MIPAS measurements (also converted into H_2SO_4 VMRs). The data are compared to MIPAS SO_2 and in a case study on two volcanic eruptions the MIPAS H_2SO_4 and SO_2 data are complemented by Chemical Transport Model (CTM) simulations. Analyses were made in terms of mass and transport patterns, to investigate the consistency of the MIPAS data sets and the evolution of volcanically emitted sulfur."

- Delete 'This paper has several purposes.'

  Thank you for this suggestion. Rather than deleting this sentence, we decided on better identifying the main purposes.

  The following changes will be made: "This paper has several purposes': (i)' we introduce a new data set of aerosol volume densities, retrieved from MIPAS measurements in Sect. 3, and '(ii)' compare the data to independent measurements of aerosols. We further study the distribution of MIPAS sulfate aerosol (as VMRs) in the period 2005 to 2012 and '(iii-b)' compare it to MIPAS SO_2. In Sect. 4 we perform (ii) a case study for two of the largest volcanic eruptions of the last decade in Northern Hemisphere mid-latitudes, which were measured by MIPAS. The volcanoes are Kasatochi (52.2° N/175° W) that erupted in August 2008, and Sarychev (48.1° N/153.2° E), which erupted in June 2009. In the case study we analyse MIPAS observations of SO_2 and stratospheric sulfate aerosol in comparison to CTM simulations, and study the sulfur mass contained in SO_2 and sulfate aerosol, together with the transport of their volcanic plumes. Finally, in Sect. 5 we draw last conclusions on the (iii) general consistency ..."

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

  The first period (Jun 2002–Mar 2004) is not considered within this study, as we aimed at investigating two of the major mid-latitudinal eruptions (Kasatochi in 2008, Sarychev in 2009) during the MIPAS measurement period (Jun 2002–Apr 2012). Volcanic eruptions during the first period only injected SO_2 masses of below 100 Tg to 10–22 km (Höpfner et al., 2015). Furthermore, the much

  longer second measurement period (Jan 2005–Apr 2012) is characterised by a better vertical and horizontal resolution due to the denser vertical and horizontal sampling. For SO_2 data are available for the entire MIPAS measurement period. Future work will be invested into the retrieval of aerosol data from the first period in order to get an aerosol data set covering the whole MIPAS lifetime.

  The revised version of the paper will include the following sentences on page 3, ln 16: "Here we concentrate on the data from the second and longer measurement period (Jan 2005–Apr 2012), as the major mid-latitudinal volcanic eruptions between 2002–2012 occurred during this period. Furthermore, this measurement period is characterised by an improved vertical resolution, especially in the altitude region of the upper tropopshere and lower stratosphere."

- Page 3: 'We analyse MIPAS observations of SO_2 and stratospheric sulphate aerosol in comparison to CTM simulations, and study the sulphur mass contained in SO_2 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.

  Thank you for pointing that out.

  In the revised version it will be made clear that the sentence is related to the case study: "'In the case study we' analyse MIPAS observations ..."

- 'Finally, in Sect. 5 we draw final conclusions on the consistency between the MIPAS SO_2 and the new MIPAS sulphate aerosol dataset,'

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

  This sentence does not only relate to the presented case study, but to the entire data sets. It includes the comparison between the two MIPAS data sets, the comparison to the in situ data, and the model data.

The sentence is changed as follows: "Finally, in Sect. 5 we draw last conclusions on the general consistency between the MIPAS SO_2 and the new MIPAS sulfate aerosol data set, in combination with our model results in the case of the two volcanic eruptions, and give a short summary of our findings."

- Page 3, ln 23: It would be good if the authors could state the time period of the SO_2 data set they are using in this study so that the reader doesn't need to look into Höpfner et al. (2015).

  The information about the time period studied here can already be found on page 3, ln 16–17 "Here we only study data from the second measurement period. During this period, from January 2005 to April 2012, ..."

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

  This will be changed in the revised version.

- Page 3, ln 30: Replace 'within' with 'between'

  This will be changed in the revised version.

- '... when due to aerosol-related sampling artefacts the total mass of SO_2 was found to be strongly underestimated (Höpfner et al., 2015).'

  Re-word to: 'when the total mass of SO_2 was found to be strongly underestimated due to aerosol-related sampling artefacts (..).'

  This will be changed in the revised version.

- 'Their study comprises a dataset of volcanically emitted SO_2 for 30 volcanic eruptions, as seen in the MIPAS measurements'

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

  To clarify that the referenced study is the study by Höpfner et al. (2015), the sentence will be changed to "The study by Höpfner et al. (2015) comprises a data set of volcanically emitted SO_2 for 30 volcanic eruptions, as observed by MIPAS.".

- 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.

  During the writing process of this paper Sect. 3 changed place. It was first included in Sect. 2, as you suggested. However, we then decided on separating the description of the new MIPAS data set from the general description of the already available data sets used in this study, and dedicateed a separate section to the new data. As one of the main purposes of the paper is to present the MIPAS aerosol data, and due to the length of the current Sect. 3, this structure seems more convenient to us.

  To make the separation into the description of available data sets in Sect. 2 and the new aerosol data set (Sect. 3) clearer, we rename Sect. 2 "2 Available observational data sets and model description" and Sect. 3 in "3 The new MIPAS aerosol data set"

- 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 ..."

  This will be changed in the revised version.

- 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.

The sentence will be updated as follows: "The latest style (Laser based counters, LPCs) of the three instrument types was used initially in 2006, became the standard Laramie instrument in 2008, and was, as an example, also flown on quasi-Lagrangian balloons in Antarctica in 2010 (Ward et al., 2014)."

- 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?

  When profiles from the two types of instruments (LPC and WPC) are available for the same day, the retrieved in situ aerosol volume density profiles show differences below 20 km, with volumes estimated from the WPC generally larger by up to a factor of two. In general the MIPAS de-biased volume density profiles show good agreement with the in situ volume density profiles retrieved from both instruments above 20 km, but below 20 km the agreement is better with the LPC. A comparison of the differences between the LPC and WPC size distribution retrievals and a judgement as to their accuracy is beyond the focus of this study. We have chosen to use the in situ instrument which permits the simplest altitude dependent de-biasing function. Before 2006 no co-incident measurements could be found to the available WPC measurements, as MIPAS measurements were relatively sparse in 2005.

  The manuscript will be changed as follows (page 4, ln 5–22): "To validate the new MIPAS aerosol data set described in Sect. 3, we use aerosol volume density profiles that were derived from in situ measurements of stratospheric aerosol

above Laramie, Wyoming (41° N, 105° W) (Deshler et al., 2003). Size resolved aerosol concentration measurements from the surface to approximately 30 km altitude were made with balloon-borne University of Wyoming optical aerosol counters. Measurements usually occurred between 6 and 9am, local time, with measurement frequency varying from monthly to bi-monthly. Data are available from 1971 to present. Over this time period three different primary instrument types were used. The latest style (Laser particle counters, LPCs) of the three instrument types was used initially in 2006, became the standard Laramie instrument in 2008, and was, as an example, also flown on quasi-Lagrangian balloons in Antarctica in 2010 (Ward et al., 2014). While the transition from the first instrument to the second was documented in Deshler et al. (2003), a similar study to compare the third Wyoming instrument with the second instrument is a work in progress. For the MIPAS validation, measurements from the second and third Wyoming instruments were available. The positive bias of MIPAS aerosol volumes from the in situ measurements was generally consistent between MIPAS and both of the Wyoming instruments above 20 km. Below 20 km the in situ measurements diverged from each other, with the second instrument indicating higher volumes than the LPC (third instrument), and at times higher than MIPAS. Based on these comparisons with both instruments the Wyoming measurements to be used were confined to those made with the LPC because it permitted the simplest altitude dependent de-biasing function for the MIPAS aerosol volume densities. The LPC measures particles with radii $>$ 0.08–4.2 μm in eight size classes."

- 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?

As this paper is not intended to study the differences between retrieved profiles following the 'old' and 'new' approach we refer to a future paper on the newly retrieved data. In the profiles tested during the preparation of the manuscript (Laramie, Wyoming; Jan 2005–Apr 2012; with co-incident MIPAS locations) in single cases large differences arose between the 'new' and 'old' profiles, while overall the approaches show similar results, particularly for the LPC. The in situ data used in this manuscript were retrieved using the new approach.

On page 4, ln 21 the following paragraph will be substituted for the current text: "Deriving geophysical quantities from the size resolved aerosol concentration measurements requires fitting a size distribution to the in situ data. In the past this has been done by fitting either a unimodal or bimodal lognormal size distribution to a subset of the measurements. The final size distribution parameters selected are those from that subset of the measurements which minimises the root mean square error when the fitted distribution is compared to all the measurements. This approach is transitioning to a new approach which modifies the nominal in situ aerosol sizes based on laboratory measurements of the aerosol counting efficiency. The counting efficiency at each size is then included in a search of the lognormal parameter space for the lognormal coefficients which minimise the error of the fitted distribution, coupled with the counting efficiency, compared to the measurements. In our study we use the volume density profiles that are de-

rived from the fitted lognormal size distributions (unimodal or bimodal, following the new retrieval approach) to the measurements. The precision of these volume estimates is the same as the old method, ±40 % (Deshler et al., 2003). The change in the way the fitting parameters are derived is the subject of a paper to be submitted soon. The impact on size distributions from the LPC measurements is not large."

- 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.

  The version of the CTM used in this work does not comprise any other than isentropic levels as possible vertical coordinates, and is therefore suitable for stratospheric studies.

  We will update the text as follows: "The Chemical Transport Model (CTM) used in our study (e.g. Sinnhuber et al.; Kiesewetter et al., 2010) is forced ... . 'The model uses isentropes as vertical coordinates.' "

- Page 4, ln 30: What are the initial OCS concentrations and where were they taken from?

  As described in Sect. 2.3, the model simulations consider as only sulphur source the volcanically emitted SO_2 from Kasatochi and Sarychev, in individual simulations. Hence, OCS is not considered, as no background SO_2 formed from OCS is included in the simulations. However, OCS is part of the newly implemented sulfur scheme, and has therefore been mentioned here.

  The following information will be included on page 5, ln 2: "... individually. 'Therefore, OCS is not considered as sulfur source in the simulations presented in this work.' "

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?

The only chemical reaction considered in this study is the reaction of SO_2 with OH, based on OH concentrations from a previous full chemistry run, and reaction rates from JPL (as described in Sect. 2.3). The model as used in this work does not include liquid and gas-phas chemistry and the implemented chemistry is applied throughout the entire model domain.

On page 4, ln 31 “In the sulfur scheme no distinction between tropospheric and stratospheric air is implemented.” will be added. On page 4, ln 5 “The sulfur released from volcanic SO_2 reacts with OH (hydroxyl radical) to form H_2SO_4. 'This is the only chemical reaction considered in the simulations presented in this study.' ”

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 SO_2 play a role here? If not, why not?

We did not consider any scavenging of SO_2 (neither H_2SO_4) in our model simulations. We expect the influence to be low in the altitude region studied here. In the tropics where the CTM reaches relatively low altitudes washout might have an impact on the sulfur distributtion. However, as can be seen in Fig. 8, in the troposphere MIPAS shows higher sulfur amounts than the CTM, even without us considering any losses due to precipitation. Furthermore, we intended to study the sulfur that enters the stratosphere, and as the MIPAS data show mostly the 'residual' SO_2' that remains in the atmosphere (above ∼10 km) after the first weeks after the eruption, we expect the influence of cloud uptake to have a minor impact, if any.

On page 5, ln 17 we add information concerning the washout by precipitation: “In our simple sulfur scheme, no scavenging of SO_2 or H_2SO_4 by clouds is considered in the model. This would be confined mostly to tropospheric altitudes and in our study region ($\geq$ 10 km) especially to tropical latitudes. Washout by precipitation might play a role there but is expected to have a minor effect on our study, as we analyse the sulfur that remains in the atmosphere (above ∼10 km) after the first weeks following the volcanic eruptions.”

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

  To clarify the difference between MIPAS/Balloon and MIPAS/Envisat, the text will be updated as follows: “In previous analyses of mid-infrared observations by MIPAS-B (the balloon-borne predecessor of the MIPAS satellite instrument; Friedl-Vallon et al., 2004) and MIPAS/Envisat (MIPAS instrument on the satellite Envisat, generally referred to as “MIPAS” throughout the present work) it has been demonstrated ...”

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

  This will be deleted in the revised version.

- 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?

  As noted in the manuscript (page 4, ln 21), the precision of the aerosol volume densities is given as ±40 %. Such error bars will be added to Figure 3 for the LPC measurements. As we only have this rough and non-variable estimate of the precision of the in situ data, it has not been considered in the calculation of the mean. LPC 2m, 1p and 3m are different instruments of the same type. These distinctions are unimportant and will be removed from the legend. All the in situ measurements will be designated as LPC.

- 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 relate to the uncertainty of the bias? This needs to be clarified.

  In Fig. 4a the standard errors of the mean profiles are shown, both for the in situ data and MIPAS data. The magnitude of these uncertainties provides information on the statistical uncertainty of the bias (difference between the mean in situ and MIPAS profile, magenta and blue profiles in Fig. 4a). To clarify what we intended to say we will include the statistical uncertainty of the bias, calculated based on the standard errors shown in Fig. 4a in Fig. 4b. This uncertainty of the difference between the in situ and MIPAS data is calculated as follows: $\sqrt{x_1^2 + x_2^2}$, with x_1 and x_2 being the standard errors at a specific altitude.

  The manuscript will be changed as follows: Page 10, ln 3: "In Fig. 4a the standard errors of the mean profiles are presented, and in Fig. 4b the statistical uncertainty of the bias (difference between the mean in situ and mean MIPAS profile) is shown." Page 10, ln 14: "In (b) the statistical uncertainty of the absolute difference between in situ and MIPAS data is shown (horizontal pink lines; square root of the sum of the 1-sigma standard errors squared for MIPAS and the in situ data)." Page 11, ln 12: "... also suits well. 'The uncertainty of the bias (Fig. 4b) at altitudes above ~17 km shows that the positive bias is not random, as the spread is low and uncertainty limits are noticeably distant from zero."

- 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.

  Thank you very much for making us realise that unfortunately information is missing on the data Fig. 4 is based on. In Fig. 3, the profiles measured on the 28.07.2011 are presented, but in Fig. 4 the profiles were calculated neglecting this day. During the work done in preparing the manuscript both calculations, including and excluding that day have been made to test how they change the mean profiles and the de-biasing, as on this day profiles show large variability between MIPAS and the in situ data at low altitudes, and strongest vertical variability of all in situ profiles ($<$ 18 km). As the de-biasing is based on altitudes from 18–30 km, it basically does not change when ex- or including that day. This is shown by this figure:

  The text will be updated as follows: Page 10, ln 1: "... mean over the profiles that were retrieved from LPC measurements, as shown in Fig. 3 (excluding the 28.07.2011) ..." Page 10, ln 3: "The profile on the 28.07.2011 shows large differences between MIPAS and the in situ data and the strongest vertical variability of all in situ profiles at low altitudes (below ~18 km), possibly due to the Nabro eruption (12 Jun 2011). Hence it is exluded from the calculation of the mean profiles shown in Fig. 4." Page 11, ln 16: "By excluding the in situ and MIPAS profiles measured on 28.07.2011 in the calculation of the mean profiles, the agreement between the measurements is improved in the altitude range below 18 km, while above this altitude changes are marginal, as can be expected from Fig. 3. The de-biasing is therefore not affected by the dismissal of the observations from this day." In the caption of Fig. 4 the following will be added: "... data in Fig. 3 '(excluding the 28.07.2011)', ..."

- 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.

The difference of the mean MIPAS profile to the mean in situ profile (both shown in Fig. 4a, as pink and blue solid lines) has been calculated, resulting in the pink solid line in Fig. 4b. The linear least squares fit to this latter profile (fit to 18–30 km, pink dashed line) represents the vertically resolved values of the de-biasing. For the de-biasing each MIPAS profile was reduced by the corresponding aerosol volume densities ($\sim$0.075 μm^{3}cm^{-3} at 10 km, $\sim$0.025 μm^{3}cm^{-3} at $\sim$27 km).

Page 11, ln 10–11 will be changed to "The de-biasing is based on the absolute differences between the aerosol volume densities of the mean MIPAS and in situ profiles (Fig. 4b, pink solid profile) at 18–30 km, where profiles show weak variability and low uncertainty of the bias. A linear least squares fit (Fig. 4b, pink dashed line) to the profile of absolute differences represents the vertically resolved values of the de-biasing, which are subtracted from each MIPAS profile during offset-correction."

- Page 13, ln 1: 'Sulphur is released from OCS mainly in the tropics at altitudes between about 25 and 35 km (Brühl 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?

  We were not refering to emissions of sulfur from OCS at tropospheric altitudes close to the surface but to stratospheric altitudes.

  In the revised version this information will be added (" 'In the stratosphere' sulfur is released ..."

- 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 eruptions according to Figure 5. What could be a cause for this increase?

  To a certain extent this signal can be distinguished in each year in which volcanic influence at these altitudes is low, most clearly visible in 2005–2007, both in the MIPAS SO_2 and aerosol data. Our CTM is not suitable for studying possible causes of these enhancements. We do not expect the signal to be caused by volcanoes, as we see it on annual basis. It might be due to strong upwelling of polluted air (anthropogenic / wild fires). However, these are speculations and are therefore not included in the manuscript.

- Figure 5: There is a strong signal at altitudes 18 to 22 km (around 2007), i.e. enhanced H_2SO_4 concentrations which cannot be seen in the SO_2 concentrations. Do the authors have an explanation for this signal in the tropics that high up?

  This signal in the aerosol data is caused by upward transport of sulfate aerosol that has been formed from volcanic SO_2 (tropical volcanoes such as Soufrière Hills and Rabaul). Vernier et al. (2011) studied SAGEII and CALIPSO data for 20° S–20° N showing the upward transport of aerosol, and a similar pattern is present in the MIPAS H_2SO_4 volume mixing ratios. The MIPAS data of SO_2 are enhanced in the tropics at comparable altitudes as the aerosol data, but only in the beginning after the eruptions, as the removal of SO_2 by chemical reaction with primarily OH is way faster than the removal of sulfate aerosol. Due to its faster removal it does not show a similar upward motion, as seen in the aerosol data.

  On page 11, ln 26 this information will be added: "... above 16 km. The aerosol is lifted upwards with time and the plumes get modulated by the Quasi-Biennial Oscillation in the tropics. A similar pattern of upward motion of the volcanic aerosol from these tropical eruptions has been seen in satellite measurements of aerosol extinction ratios (Vernier et al., 2011)."

- 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.

  In the CTM we use a constant aerosol radius to determine the terminal fall velocity, intending to simulate sedimentation with a constant average settling velocity that corresponds to aerosol of different radii.

  We will name the simulation radius "effective sedimentation / settling radius" throughout the paper and add the following information on page 5, ln 17: "In the atmosphere the radius of sulfate aerosol varies (Deshler et al., 2003 and 2008). Nevertheless, for simplification we use a constant "effective sedimentation radius" to determine the terminal fall velocity, which we consider to be the average settling speed of aerosol particles of different radii."

- Page 14, ln 9: 'Good accordance between the modelled and measured SO_2 masses is essential to test, ...'

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

  The sentence will be changed as follows: "As we intend to test if the measured aerosol is quantitatively and qualitatively consistent with its measured precursor by comparison with modelled sulfate aerosol, a good agreement between the modelled and measured SO_2 masses is essential."

- Page 14, ln 11: 'In Table 1, SO_2 masses for three altitude regions ...' Include 'injected SO_2 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.

  Your suggestion will be included in the revised version. The injected SO_2 amounts have been chosen from a sensitivity study with differing injected masses (partly

  based on masses as given by Höpfner et al., 2015), with the intention to achieve good agreement between the modelled and measured SO_2 (agreement starting approx. one month after the respective eruption; comparison of masses as in Fig. 6). The presented SO_2 masses resulted in the best agreement.

  This will be clarified on page 14, ln 14: "Simulations have been made with varying injected SO_2 masses and upper injection altitude limits, intending to achieve good agreement between the modelled and measured SO_2 masses (comparisons as in Fig. 6). The data presented here resulted in the best agreement, with comparisons starting approximately one month after the respective eruption (explanation in the following)."

- Page 14, ln 12: 'The simulations result in good agreement between measured and modelled SO_2.'

  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.

  We are referring to the simulations as presented in Fig. 6 (orange and blue lines). The simulations were made with a time step of 30 min, started on the last day of the month preceeding the eruption, espectively, and covered 365 d. As the background for SO_2 and H_2SO_4 has been set to be zero throughout the entire model domain, and the model is driven by ERA-Interim data that are read in every 6 h, no real spin up time is needed. As the mentioned sentence is related to a Figure that is only described later (Fig. 6), it will be deleted here.

  Instead, on page 17, ln 18 we add the following sentence: "After this first month, the simulated SO_2 agrees well with the measurements by construction." In Sect. 2.3 "Chemical Transport Model" the following information will be added (page 5, ln 17): "The model is run for 365 d per simulation, with a time step of 30 min and tracer fields are written out daily at 12 UTC. For the eruption of

Kasatochi (7 Aug 2008) the individual runs are started on the 31 Jul 2008, and for the eruption of Sarychev (12 Jun 2009) all runs are started on the 31 May 2009. As the initial tracer fields are set to zero and the model is driven by ERA-Interim reanalysis data, which are updated every six hours, no long spin up time is needed. Per volcano four simulations were made that differ concerning the particle size of sulfate aerosol. Simulations were made with constant aerosol radii of 0.1, 0.5 and 1 μm, and without sedimentation."

- Page 14, ln 14: 'This method was applied as in the first month after the eruption MIPAS underestimates the SO_2 (Höpfner 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 Höpfner et al., (2015). Why is it important to know how Höpfner et al. derived the mass? How did the authors derive the SO_2 mass used for their study? Could the authors please clarify why they didn't use the same values as Höpfner et al. and how they derived their values for SO_2 mass injected by the two volcanoes? And why do they not provide any uncertainties on their values?

  For parts of your questions we refer to a previous question where we answered how the injected SO_2 masses were derived in our study. The method mentioned here was applied in the study by Höpfner et al. (2015), not in our study. However, as the injected SO_2 masses are partly based on the study by Höpfner et al. (2015) we think that is is valuable to include this information in our manuscript. For both volcanoes the simulations resulted in better agreement with the measurements when injecting less SO_2, compared to the masses given by Höpfner et al. (2015). For Kasatochi the masses by Höpfner et al. (2015) minus their given uncertainties are used, which are relatively large, due to the method they applied to derive the masses. The number of simulations we made was not sufficiently large to calculate uncertainties for the presented SO_2 masses.

  The manuscript will be changed as follows: "They applied this method as in the

  first month after the eruption MIPAS underestimates the SO_2 (Höpfner et al., 2015). Their method results in relatively large error bars that depend on the time period the fit is based on (Höpfner et al., 2015; presented also in Table 1)." On page 14, ln 14 the following information will be added: "Due to the limited number of simulations no uncertainties are given for the presented SO_2 masses. The main part of SO_2 ..."

- 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?

  This was only an internal label indicating "no sedimentation" - we will correct this.

  The statement on page 16, ln 4–5 will be corrected to: "For the CTM four simulations for H_2SO_4(liq) are shown with different effective sedimentation radii (0.1, 0.5, and 1 μm), and without sedimentation." On page 15, ln 8–9 the statement will be changed to: "The simulations differ concerning the implemented sedimentation (no sedimentation and effective sedimentation radii of 0.1, 0.5, and 1 μm)."

  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 SO_2 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.

  No additional SO_2 has been injected in the model due to the eruption of Redoubt. It is mentioned here, as it can be seen in the MIPAS measurements.

  To clarify why it is indicated in Fig. 6, on page 15, ln 11 the following information will be added: "In Fig. 6, the eruption times of Kasatochi (7 Aug 2008) and Sarychev (12 Jun 2009) are indicated. Additionally, the eruption time of Redoubt (23 Mar 2009) is marked, as this eruption produces a signal in the measurements. It is not included in the simulations, however."

What in the CTM causes SO_2 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.

The possible reasons for loss of SO_2 in the model from a confined altitude and latitude region are due to chemical loss or to vertical or horizontal transport out of the region. As noted in Sect. 2.3, the vertical transport is calculated based on ERA-Interim heating rates, the horizontal transport based on ERA-Interim wind fields, and for the chemical lifetime the concentration of OH and the ambient temperature and pressure are important. We did not study which process dominates the removal in the different altitude regions shown in Fig. 6.

On page 15, ln 11 the follwing information will be added: “In the model the two species can be removed from one confined altitude and latitude region due to transport (advection: SO_2, H_2SO_4; sedimentation: H_2SO_4) or chemical loss (SO_2).”

The possible reasons for loss of SO_2 in the model from a confined altitude and latitude region are due to chemical loss or to vertical or horizontal transport out of the region. Loss of sulphate aerosol in the model is possible due to sedimentation and advective transport. The sum of SO_2 and sulphate aerosol neglects the chemical loss and comparisons between simulations with and without sedimentation show the influence of sedimentation on the removal. A comparison of Fig. 6 (a) and (d) reveals a fast decay of the total sulfur in the case of Kasatochi, which is mostly connected to advection (removal in the Brewer-Dobson circulation), as the differences between the simulations with and without sedimentation are low and the fast decay is present both in the curve of SO_2 and total sulfur. Only after some months the effect of sedimentation increases, when sulfur from above reaches this altitude region (second peak). In the case of Sarychev the curve of SO_2 and total sulfur are rather different, as especially the sulfate aerosol is supplied by aerosol that is transported downwards from above. This can also

be seen in Fig. 7.

Can the authors explain why for a radius of 1 μm the H_2SO_4 loss is levelling off earlier (around November) between 18.5 and 22.5 km than compared to the other altitude ranges?

In the uppermost altitude range only little aerosol is present, and aerosol with a radius of 1 μm settles relatively fast, compared to smaller particles. Furthermore, at higher altitudes sedimentation velocities are higher. Therefore the relatively little excess amount of aerosol is removed rapidly.

On page 18, ln 8 the follwing information will be added: “... can be expected. Model simulations show that compared to 10.5–18.5 km and compared to small particles, the bigger particles level out faster in the uppermost altitude range studied here. Reasons for this faster removal of the volcanic aerosol are that only little aerosol is injected in the altitude region 18.5–22.5 km, that bigger particles settle faster, and that settling velocities rise with increasing altitude due to the corresponding decrease in air density.”

What is causing the second peak (around Dec) in H_2SO_4 between 10.5 and 14.5 km? This peak is also seen in the MIPAS measurements but shifted by half a month. The peak in SO_2 as modelled by the CTM is shifted compared to the peak in MIPAS SO_2. Can the authors explain why that is?

This second peak is caused by downward transport of sulfur that has been emitted by the eruption of Kasatochi. It is described on page 19, ln 1–8. The time shift has not been studied in detail, but is assumed to be caused by model uncertainties and sampling artefacts, as in this altitude region both the measurements and model data are sparse.

We will mention the second peak on page 18, ln 25: “A peak can be seen in the measured and modelled sulfur dioxide and sulfuric acid masses in November / December 2008 (Fig. 6) in the lowermost altitude region (10.5–14.5 km). This peak is caused by downward transport of sulfur in the extra-tropics that has been emitted by the eruption of Kasatochi. In the following section (Sect. 4.2) more details are given on this transport pattern.”

- 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.

  This will be changed in the revised version.

- 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 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.

  It is true that we need to clarify better why we say that the no-sedimentation run already compares well to the measurements. We are refering to the long-term decay of the sulfur mass. In the beginning sedimentation has a strong influence on the absolute amount of sulfur and the impact on the sulfur that still remains in the altitude ranges after a long period of time is clearly visible, but the long-term decay is quite similar in all simulations. The long-term removal is not dominated by sedimentation but by transport in the Brewer-Dobson circulation. Furthermore, the upper altitude range where sedimentation is fastest has no strong impact on the absolute sulfur mass and its removal as only a minor amount of sulfur is found at these altitudes.

  On page 1, ln 20 will be changed to: “While sedimentation of sulfate aerosol plays a role, we find that the long-term decay of stratospheric sulfur after these volcanic eruptions at mid-latitudes is controlled mainly by transport in the Brewer-Dobson

circulation.” On page 18, ln 19 the following will be added: “This becomes obvious when comparing the ’long-term removal of’ total modelled sulfur ...” On page 18, ln 23–24 this information will be added: “... the modelled ’decay of’ sulfur mass without sedimentation already compares rather well with the measured ’decay of’ sulfur mass.” On page 24, ln 6 will be changed to “While sedimentation of sulfate aerosol does play a role, the ’decay’ of sulfur in the mid-latitude lower stratosphere ...”

- Figure 7: Again why is the Redoubt eruption included in this Figure.

  The eruption of Redoubt is indicated, as the MIPAS data show a signal caused by this eruption.

  The caption of Fig. 7 will be updated as follows (page 19, ln 16): “Indicated are the days of the eruptions of the volcanoes Kasatochi in Aug 2008, Redoubt in Mar 2009, and Sarychev in Jun 2009, which were observed by MIPAS. Redoubt is not considered in the model.” In the captions of Fig. 8 and 9 similar information is added.

  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.

  On page 5, ln 17 the following information will be added: “The model is run for 365 d per simulation, with a time step of 30 min and tracer fields are written out daily at 12 UTC. For the eruption of Kasatochi (7 Aug 2008) the individual runs are started on the 31 Jul 2008, respectively, and for the eruption of Sarychev (12 Jun 2009) all runs are started on the 31 May 2009. As the initial tracer fields are set to zero and the model is driven by ERA-Interim reanalysis data, which are updated every six hours, no long spin up time is needed.”

  Why is the pattern for H_2SO_4 (CTM simulation) for the Kasatochi eruption so different from the Sarychev eruption? For the Kasatochi eruption the H_2SO_4 concentrations are lower than for the Sarychev eruptions although similar amounts of SO_2 were injected?

One big difference between the eruptions of Kasatochi and Sarychev can already be seen in Fig. 6 and Table 1. In the case of Kasatochi a relatively large part of the SO_2 is injected at altitudes below 14 km (518 Gg or 77% of the injected SO_2) and transported downwards and out of the altitude region we study in the present work relatively fast. Therefore it is not reflected in the aerosol loading. In the case of Sarychev more of SO_2 is injected into the altitude region above 14 km (367 Gg or 48% of the injected SO_2). Hence, more SO_2 is available after the eruption of Sarychev to be converted into sulfate aerosol and to stay in the altitude range studied here for a relatively long period of time.

On page 19, ln 11 this information will be added: “Parts of the differences between the transport patterns after the eruptions arise from the injected SO_2 masses. In the case of Kasatochi the main part of SO_2 was injected to altitudes below 14 km (518 Gg or 77 % of the injected SO_2). It is transported downwards and out of the region studied here relatively fast and therefore only a minor part is reflected in the aerosol loading. In the case of the Sarychev eruption almost half of the SO_2 (367 Gg or 48 % of the injected SO_2) is injected into the altitude region above 14 km. It is available for conversion into sulfate aerosol for a longer period of time, as can be seen in the higher H_2SO_4 volume mixing ratios after the eruption of Sarychev, compared to Kasatochi.”

How much of the simulated Kasatochi ’double-plume’ is made due to the choice of the SO_2 mass being injected at different altitude levels?

Neither in a CTM simulation with the SO_2 mass from Kasatochi, which erupted on the 7 Aug 2008 at 52.2° N/175.5° W, injected on the day and at the location of the Sarychev eruption (12 Jun 2009, 48.1° N/153.2° E), nor in a CTM simulation with the Kasatochi SO_2 mass injected at the location of Sarychev (7 Aug 2008, 48.1° N/153.2° E), a comparable separation into an upper and lower part of the plume is simulated. When the SO_2 mass from Sarychev is injected on the day

and at the location of the Kasatochi eruption (7 Aug 2008, 52.2° N/175.5° W) no strong separation is simulated either. These CTM simulations therefore suggest that the pattern of the ’double-plume’ is caused by the meteorological situation after the eruption in combination with the vertical distribution of injected SO_2.

On page 19, ln 11 this information will be added: “Model simulations with ’switched’ SO_2 masses (mass of Kasatochi injected on the day and at the location of Sarychev, and vice versa), and a simulation with the SO_2 mass from the Kasatochi eruption injected at the location of Sarychev, reveal that the ’double-plume’ that has been observed after the eruption of Kasatochi results from the combination of the vertical distribution of injected SO_2 masses and the prevailing transport after the 7 Aug 2008, the eruption date of Kasatochi, in the model driven by wind fields and heating rates. Neither of the simulations results in a comparable separation into an upper and lower part of the plume.”

The injection heights of SO_2 for the Sarychev eruption seem not appropriate when compared with the measurements. The model simulations show high SO_2 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?

When keeping in mind that MIPAS has problems in detecting SO_2 up to approx. one month after the eruption, we do not see such a strong mismatch between the altitude range covered by the measured and modelled sulfur dioxide after the eruption of Sarychev (right plume in each plot in Fig. 7). Concerning the injection height and the impact on our results, no extensive sensitivity assessment has been done. However, we made simulations with varying upper injection limits and concluded that 19 km was resulting in reasonable agreement between the measurements and simulations.

- 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?

The concentrations are reduced during the downward transport of the upper part of the plume, and not the bulk but some parts of the plume reach 10 km after several months. It is true that only very few sulfur reaches 10 km.

This part of the sentence will be replaced by: "... circulation. During the descent the sulfur concentrations are reduced and some parts of the sulfur reach 10–12 km after several months."

- Figure 8: The CTM model shows about 1 month lag before SO_2/H_2SO_4 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?

As can be seen in Fig. 5 and has been noted in the manuscript, the MIPAS SO_2 and H_2SO_4 data show enhancements in the tropics at altitudes of about 14–16 km during the entire measurement period (with varying intensities), which are supposed to only partly be connected to volcanic eruptions. A certain influence of elevated aerosol values due to cirrus clouds that have not been captured by the ice-filter is possible. We cannot quantify from the observations to which extent the enhancements seen in the measurements in Fig. 8 are caused by the eruptions of Kasatochi and Sarychev. It is clear that parts of the sulfur that has been observed from November 2008 to ∼February 2009 has been injected into the atmosphere by the eruption of Dalafilla (Nov 2008).

On page 20, ln 9, the following information will be added: "The relatively high values in the measurements at around 13–16 km have already been noted in Fig. 5 and are supposed to only partly be connected to volcanic eruptions. A

certain influence of elevated retrieved aerosol values due to cirrus clouds that have not been captured by the ice-filter (Sect. 3.1) is possible. To which extent the observed enhancements in the measurements (Fig. 8a–c) are caused by the eruptions of Kasatochi and Sarychev is not clear. In the case of Kasatochi model simulations suggest that enhancements are confined primarily to altitudes above approximately 16 km. Additionally to the tropical enhancements at 13–16 km, the eruption of Dalafilla in November 2008 overlays with the observed sulfur that has been emitted by Kasatochi. The CTM simulations of Sarychev indicate that sulfur observed at altitudes as low as 12 km can be attributed to the volcanic eruption."

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 eruption is seen in the tropics) seems to be reduced for the Sarychev eruption? The model seems to overestimate the peak amount of SO_2+H_2SO_4 compared to the observations. Any explanation for that given that the model agrees with the peak values of measurements better between 30 and 90N?

Comparisons of daily global horizontal distributions of the measurements and model results show that the differences between the measurements and simulations seen in Fig. 8 are partly due to sampling artefacts and partly due to a different horizontal extent of the volcanic plumes. In some regions where the model data show enhancements, measurements were filtered out due to clouds (ice, water, ash), especially in the tropics (SO_2: up to around 14/15 km and in the region of the Asian monsoon up to around 16/17 km; aerosol: primarily up to around 18/19 km and in the region of the Asian monsoon up to about 20 km). This might produce a low bias in the zonal mean of the measurements, compared to the CTM data that covers the entire globe (above 13 km). Additionally, the horizontal extension of the plumes is not in perfect agreement between the measurements and observations, provoking further differences between the zonal mean volume mixing ratios. In the case of the Sarychev eruption a relatively strong meridional transport of the sulfur is simulated already in the first days after the eruption. Simulated SO_2 reaches 15° N 8 d after the eruption at 14 km and 16 km, and 12 d after the eruption at 18 km (with VMR $>$ 1,500 pptv). This strong meridional tranpsport is not reflected in the measurements – where available – leading to the time lag and higher sulfur content in the tropics in the modelled data.

On page 20, ln 9, the following information will be added: “Differences between the presented zonally averaged measurements and model results arise partly from the fact that MIPAS measurements are not uniformly distributed and data were filtered, and due to sparse data coverage in the case of the CTM up to an altitude of 12–13 km. Data are partly missing in relatively large areas, which may lead to biased zonal means. In the measurements, for SO_2 data are missing particularly in the tropics at altitudes below about 15/16 km and at higher altitudes (up to ∼17 km) in the region of the Asian Summer Monsoon. In the case of measured sulfate aerosol data are filtered especially in the tropics at altitudes up to about 18/19 km and in the region of the Asian Summer Monsoon (up to ∼20 km) and in polar regions entire profiles were filtered out due to PSCs. In the case of the CTM data coverage is reduced up to an altitude of 13 km. Especially after the eruption of Sarychev a higher sulfur content is simulated in the tropics compared to the measurements (Fig. 8) and enhancements are seen few days after the eruption. This results from a strong modelled meridional transport of SO_2 after this eruption. At about 12–16 km altitude the injected SO_2 reaches 15° N 7–8 d after the eruption. This strong southward transport early after the eruption is not reflected in the measurements, which are, however, partly missing in the tropics due to filtering.”

- Figure 9: For the CTM simulations, it looks like SO_2 is immediately enhanced at lower latitudes (down to 15deg N) at the time of the eruption, especially for the Sarychev eruption. Why is that? SO_2 is much more confined to the NH in the

observations.

Due to the prescribed wind fields during the first days after the eruption, in the model the Sarychev plume is spread relatively fast in meridional directions. At that time only few MIPAS measurements show high values, and not that far South. Also after a longer period, the measurements (where available and not filtered out) do generally show less enhancements in the tropics than the CTM. The differences might be caused by model uncertainties or inaccuracies in the prescribed wind fields. Difficulties with the measurements / retrieved profiles are another possible reason for the differences. After the eruption of Kasatochi less of a latitudinal spread is simulated and the plume is rather compact in the beginning. It takes about 16 days for the plume to reach 15° N. At that time the MIPAS SO_2 already shows a relatively strong signal produced by the volcano, that extends to these tropical latitudes.

How is the SO_2 emitted by the volcanoes injected into the model? The authors state that ’SO_2 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 SO_2 was uniformly distributed between 10 and 14 km (just looking at the time of the eruption, realizing that things will change after some time)?

In the model the SO_2 is injected on the day of the eruption at 12 UTC, into the column of grid boxes that includes the location of the volcano. The mass for a given altitude range has been distributed equally to the air mass contained in the grid boxes for which the centres lie in this altitude range, excluding the lowermost (boundary) level. The differences in the meteorological situation / wind fields and in the injected sulfur dioxide masses result in the differences between the simulations of Kasatochi and Sarychev. The vertical distribution of grid boxes is rather similar at the eruption times and locations of Kasatochi and Sarychev, therefore, this can be excluded as explanation for the differences.

I find the differences in the latitudinal distribution of SO_2 between the observations and the model interesting and would like the authors to comment on that.

As noted before and included in the manuscript, sampling artefacts and differences in the horizontal extent / location of the plumes cause disagreement between the zonal mean values calculated from simulations and observations.

- Page 22, ln 20: '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 SO_2. Do the authors have any explanation for the 'leak' towards the equator in the model, especially in July/Aug after the Sarychev eruption?

  As noted before sampling artefacts and differences in the horizontal extent / location of the plumes cause disagreement between the zonal mean values calculated from simulations and observations. In the case of the Sarychev eruption a strong meridional transport is simulated early after the eruption, which cannot be proved by comparisons to the MIPAS measurements. Partly due to missing data in the region of high modelled sulfur content and partly due to elevated simulated sulfur amounts in regions of low measured VMRs.

- 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?

  This sentence refers to the southward transport of sulfur.

  For clarification the manuscript will be changed to: "At an altitude of about 18 km in the case of Kasatochi and $\sim$16 km in the case of Sarychev, and at altitudes above, a southward transport of sulfur is noticed (Fig. 9 and 10)."

- Figure 10: Enhanced H_2SO_4 concentrations reach further South in the model simulations than the observations show (at 18 km). In the observations H_2SO_4 is more confined to latitudes between 15 and 60deg N while for the model simulations, enhanced H_2SO_4 concentrations reach the equator. Why the difference?

  In the tropics at an altitude of 18 km the data coverage for MIPAS H_2SO_4 is relatively sparse. In the case of Sarychev it is not clear if a better data coverage would improve the agreement or if it is a 'real' difference between the measurements and the model results, which show enhancements that reach the equator. After the eruption of Kasatochi the modelled H_2SO_4 is transported southwards but elevated values do not reach that far towards the equator. Therefore a possible influence of filtered MIPAS data on the differences is weaker.

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

  This sentence will be changed to: "Compared to Sarychev, the southward transport of the Kasatochi eruption plume is weaker and initiates at higher altitudes. This might be explained by the eruption of Kasatochi having been later during the monsoon season, resulting in a shorter time period of enhanced southward transport induced by the Asian summer monsoon."

- Page 23, ln 20: 'In this study a new dataset of MIPAS/Envisat global aerosol volume densities and liquid-phase H_2SO_4 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.

We are actually talking about one data set of aerosol volume densities that has been converted into H_2SO_4 volume mixing ratios.

The sentence will be changed as follows: "In this study a new data set of MIPAS/Envisat global aerosol volume densities, also converted into liquid-phase H_2SO_4 volume mixing ratios, is presented for ..."

- Page 24: 'The new H_2SO_4 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 SO_2 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.

  On page 24, ln 5 we will add this information: "The new estimates are 677 Gg SO_2 in the case of Kasatochi and 768 Gg SO_2 in the case of Sarychev that were injected into the altitude range 10–19 km."

- 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.

  The OH levels have been derived in a CTM full chemistry run (2003–2006), without any feedback between the sulfur species and the OH concentrations.

  For clarification the following will be added at page 24, ln 6: "... under OH background conditions (modelled OH climatology without any feedbacks between sulfur species and OH concentrations)."

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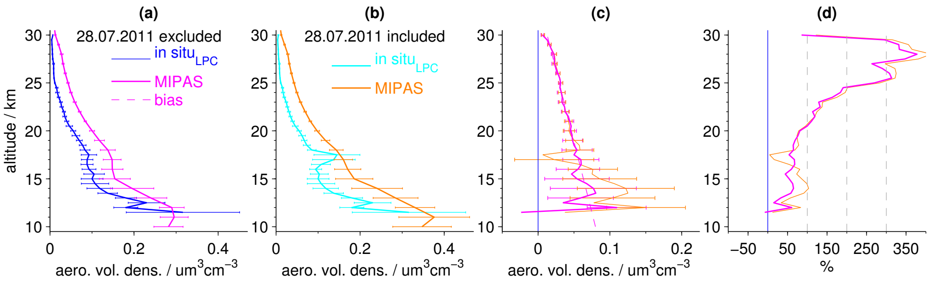
(a)
(b)
(c)
(d)
28.07.2011 excluded
in situ$_{LPC}$
MIPAS
bias
28.07.2011 included
in situ$_{LPC}$
MIPAS
altitude / km
30
25
20
15
10
0
0.2
0.4
0.1
−50
50
150
250
350
aero. vol. dens. / um^{3}cm^{-3}
%

Fig. 1.

MIPAS observations of volcanic sulfate aerosol and sulfur dioxide in the stratosphere

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Abstract

Volcanic eruptions can increase the stratospheric sulfur ~~content~~ loading by orders of magnitude above the background level and are the most important source of variability ~~of~~ for stratospheric sulfur ~~loading~~. We present a set of vertical profiles of sulfate aerosol volume densities and derived liquid-phase H_2SO_4 (sulfuric acid) mole-fractions for 2005–2012, retrieved from infrared limb emission measurements by the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) on board ~~of~~ the Environmental Satellite (Envisat). ~~The MIPAS aerosol data set has been corrected for an altitude-dependent bias by comparison with balloon-borne in situ aerosol measurements at Laramie, Wyoming~~. In comparison to balloon-borne in situ measurements of aerosol at Laramie, Wyoming, the MIPAS aerosol data have a positive bias that has been corrected, based on the difference to the in situ data. ~~The MIPAS data of stratospheric sulfate aerosol are linked to MIPAS observations of sulfur dioxide (SO_2) with the help of Chemical Transport Model (CTM) simulations.~~ We investigate the production of stratospheric sulfate aerosol ~~and its fate~~ from volcanically emitted SO_2 for two ~~volcanic~~ case studies: the eruptions of Kasatochi in 2008 and Sarychev in 2009, which both occurred in the Northern Hemisphere mid-latitudes during boreal summer. With the help of Chemical Transport Model (CTM) simulations for the two volcanic eruptions, we ~~We~~ show that the MIPAS sulfate aerosol and SO_2 data are qualitatively and quantitatively consistent to each other. Further, we demonstrate that the lifetime of SO_2 is well described by its oxidation by hydroxyl radicals (OH). While sedimentation of ~~the~~ sulfate aerosol plays a role, we find that the long-term decay of stratospheric sulfur after these volcanic eruptions at mid-latitudes is mainly controlled by transport in the Brewer–Dobson circulation ~~dominant mechanism controlling the stratospheric lifetime of sulfur after these volcanic eruptions at mid-latitudes is transport in the Brewer–Dobson circulation~~. Sulfur emitted by the two mid-latitude volcanoes resides mostly north of 30° N at altitudes of ~10–16 km, while at higher altitudes (~18–22 km) part of the volcanic sulfur is transported towards the equator where it is lifted into the stratospheric 'overworld' and can further be transported into both hemispheres.

1 Introduction

Aerosol particles are omnipresent in the atmosphere and can affect climate, air quality, and atmospheric chemistry. In the stratosphere~~,~~ aerosol particles are mainly composed of sulfuric acid (H_2SO_4) and water (H_2O) (Kremser et al., 2016; Thomason and Peter, 2006), though organic material is demonstrated to also play a significant role in the upper troposphere and lower stratosphere (Yu et al., 2016; Murphy et al., 2014). Stratospheric sulfate aerosol has the potential to directly cool surface temperatures by backscattering parts of the incoming solar radiation. Estimates of the amount of stratospheric aerosol~~,~~ and their evolution with time~~,~~ are therefore important for climate change modelling studies. Increased interest in stratospheric sulfate aerosol is also connected to its potential use in climate engineering schemes (e.g. Niemeier and Timmreck, 2015; Rasch et al., 2008). The negative radiative effect of stratospheric aerosol (Andersson et al., 2015; Brühl et al., 2015; Ridley et al., 2014; Santer et al., 2014; Solomon et al., 2011; Vernier et al., 2011) is discussed to be one of the causes for the global warming 'hiatus' experienced during the first decade of this century (Haywood et al., 2013 ~~2014~~; Santer et al., 2014; Fyfe et al., 2013). Hofmann et al. (2009) observed an increase in ~~of~~ stratospheric aerosol load and ~~speculated~~ suggested that this ~~is~~ was due to anthropogenic emissions. Newer studies, however, show that this increase is likely to be connected to a number of small and medium sized volcanic eruptions especially in the tropics ~~this increase to be connected more likely to a series of smaller and medium sized tropical volcanic eruptions~~ (e.g. Neely et al., 2013; Vernier et al., 2011). ~~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.~~ ~~These~~ During the last decade several volcanoes directly injected sulfur up to 20 km into the stratosphere (Vernier et al., 2011). ~~Ridley et al. (2014 and Andersson et al. (2015) 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.~~ Ridley et al. (2014) and Andersson et al. (2015) emphasise the importance of volcanic aerosol in the lowermost stratosphere at mid- and high-latitudes on the total volcanic aerosol forcing during the last decade. Their studies show that stratospheric altitudes below ~15 km (380 K isentrope), which are not represented in many of the aerosol data sets, need to be taken into consideration when studying the global radiative forcing generated by volcanic eruptions in the extra-tropics. ~~Understanding of stratospheric sulfur, 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).~~

The main source gases of stratospheric sulfate aerosol during background / non-volcanic conditions are sulfur dioxide (SO_2) and carbonyl sulfide (OCS). Due to the longer lifetime of OCS compared to SO_2, carbonyl sulfide has a relatively high flux across the tropical tropopause layer (TTL), its main entry pathway into the stratosphere. Crutzen (1976) first stated the essential role of OCS for stratospheric aerosol. ~~E.g.,~~ Chin and Davis (1995), Thomason and Peter (2006), Brühl et al. (2012), and Sheng et al. (2015), agree on a major contribution of OCS to stratospheric sulfate aerosol. However, ~~its exact contribution to stratospheric aerosol~~ the magnitude to which OCS contributes to the stratospheric aerosol loading during background conditions is still ~~in~~ under discussion. By emitting SO_2, volcanic eruptions are the dominant source for stratospheric SO_2 (direct) and sulfate aerosol (indirect) under non-background conditions, and cause most of the variability in the stratospheric sulfur loading. ~~During volcanically perturbed times volcanically emitted SO_2 is the dominant source for stratospheric sulfate aerosol and causes most of the variability in the stratospheric sulfur loading~~ content~~. Some eruptions directly injected SO_2 into the stratosphere (e.g. Vernier et al., 2011). In volcanic emissions, SO_2 is the third most abundant emitted gas, after water vapour and carbon dioxide (von Glasow et al., 2009).~~

~~To study stratospheric sulfur a combination of observations and model simulations is useful.~~ When analysing the vertical extent of ~~the~~ SO_2 and aerosol plumes after volcanic eruptions and their transport at different altitudes, vertically resolved observations are needed. These are available from satellite limb measurements, such as the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS), an instrument that was operational on Envisat (Environmental Satellite). From 2002 to 2012 the instrument delivered limb emission measurements in the infrared spectral region. From MIPAS several ~~datasets~~ data sets of trace gas species that are relevant to study the stratospheric sulfur ~~content~~ loading are already available. These are volume mixing ratios (VMRs) of OCS (Glatthor et al., 2015 and 2017), and SO_2 (Höpfner et al., 2013 and 2015). ~~Here we present an additional data set of sulfate aerosol from MIPAS, and combine the MIPAS SO_2 and liquid-phase H_2SO_4 measurements with Chemical Transport Model (CTM) simulations to analyse the consistency of the two data sets, and the evolution of volcanically emitted sulfur.~~ Here, we present a new data set of sulfate aerosol volume densities (AVDs) retrieved from MIPAS measurements (also converted into H_2SO_4 VMRs). The data are compared to MIPAS SO_2 and in a case study on two volcanic eruptions the MIPAS H_2SO_4 and SO_2 data are complemented by Chemical Transport Model (CTM) simulations. Analyses were made in terms of mass and transport patterns, to investigate the consistency of the MIPAS data sets and the evolution of volcanically emitted sulfur.

In Sect. 2 we first provide basic information on MIPAS, the MIPAS SO_2 ~~dataset~~ data set, and balloon-borne in situ data of aerosol volume densities used in this study. This is followed by a short description of the CTM and our model implementations. This paper has several purposes~~.~~ : (i) we ~~We~~ introduce a new ~~aerosol dataset~~ data set of aerosol volume densities, retrieved from MIPAS measurements in Sect. 3, and compare the data to independent measurements of aerosols. We further study the distribution of MIPAS sulfate aerosol (as VMRs) in the period 2005 to 2012 and compare it to MIPAS SO_2. In Sect. 4 we perform (ii) a case study for two of the largest volcanic eruptions of the last decade in Northern Hemisphere mid-latitudes, which were measured by MIPAS. The volcanoes are Kasatochi (52.2° N/175.5° W) that erupted in August 2008, and Sarychev (48.1° N/153.2° E), which erupted in June 2009. In the case study we ~~We~~ analyse MIPAS observations of SO_2 and stratospheric sulfate aerosol in comparison to CTM simulations, and study the sulfur mass contained in SO_2 and sulfate aerosol, together with the transport of their volcanic plumes. Finally, in Sect. 5 we draw ~~final~~ last conclusions on the (iii) general consistency between the MIPAS SO_2 and the new MIPAS sulfate aerosol ~~dataset~~ data set, in combination with our model results in the case of the two volcanic eruptions, and give a short summary of our findings.

2 ~~Data sets and methods~~ Available observational data sets and model description

2.1 MIPAS

2.1.1 Instrument

MIPAS (Fischer et al., 2008) is an infrared (IR) limb emission sounder that operated on ESAs (European Space Agency) satellite Envisat. The Fourier transform spectrometer measured high-resolution spectra emitted by the constituents of the atmosphere in the thermal IR, in the region 685 to 2,410 cm^{-1} (ESA, 2000). The instrument operated from July 2002 to April 2012, separated in two measurement periods. Here we concentrate on the data from the second and longer measurement period (January 2005–April 2012), as the major mid-latitudinal volcanic eruptions between 2002–2012 occurred during this period. Furthermore, this measurement period is characterised by an improved vertical resolution, especially in the altitude region of the upper troposphere and lower stratosphere. ~~Here we only study data from the second measurement period.~~ During this period~~, from January 2005 to April 2012,~~ radiance profiles from 7 to 72 km altitude were measured, with an ~~unapodized~~ unapodised spectral resolution of 0.0625 cm^{-1}, a latitudinal distance of 420 km between two subsequent limb scans, and a vertical sampling step of 1.5 km in the upper troposphere / lower stratosphere region. Installed on a sun-synchronous polar orbiting satellite, at an altitude of about 800 km, MIPAS delivered data at around 10am and 10pm, local time. For the retrieval of sulfate aerosol volume densities, described in this paper, MIPAS level 1b calibrated radiances version 5 were used, as provided by ESA.

2.1.2 The SO_2 ~~dataset~~ data set

In this study~~,~~ we use the MIPAS SO_2 ~~dataset~~ data set as described by Höpfner et al. (2015). Error estimations and a validation of the SO_2 ~~dataset~~ data set by comparison with satellite data from the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) and other available SO_2 observations are provided by Höpfner et al. (2015). Single SO_2 profiles of this ~~dataset~~ data set have a total estimated error of around 70–100 pptv and a vertical resolution of 3–5 km. The MIPAS data are shown to be consistent with independent measurements from several aircraft campaigns within ± 50 pptv. With respect to satellite-borne data from ACE-FTS, MIPAS SO_2 shows a bias ~~of~~ between -20 to 50 pptv during volcanically perturbed periods that differs with altitude~~, which is a reasonably small bias, given the natural variability of volcanic SO_2~~. For background conditions this bias lies ~~within~~ between -10 to 20 pptv in the altitude region 10–20 km. For the analysis of volcanically enhanced periods it is necessary to stress complications of the MIPAS SO_2 data up to a few weeks directly after the eruption when the total mass of SO_2 was found to be strongly underestimated, especially due to aerosol-related sampling artefacts (Höpfner et al., 2015) ~~when due to aerosol-related sampling artefacts the total mass of SO_2 was found to be strongly underestimated (Höpfner et al., 2015)~~. The study by Höpfner et al. (2015) ~~Their study~~ comprises a ~~dataset~~ data set of volcanically emitted SO_2 masses for 30 volcanic eruptions, as ~~seen in the~~ observed by MIPAS ~~measurements~~.

2.2 Aerosol in situ measurements from Laramie, Wyoming

~~To validate the new MIPAS aerosol dataset, we use aerosol volume density profiles that were derived from in situ measurements of stratospheric aerosol (Deshler et al., 2003) above Laramie, Wyoming (41° N, 105° W). 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. Measurements usually occurred between 6 and 9am, local time, with the measurement frequency varying from monthly to bi-monthly. Data are available from 1971 to present. Over this time period three different primary instrument types were used. 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). 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.~~ To validate the new MIPAS aerosol data set described in Sect. 3, we use aerosol volume density profiles that were derived from in situ measurements of stratospheric aerosol above Laramie, Wyoming (41° N, 105° W) (Deshler et al., 2003). Size resolved aerosol concentration measurements from the surface to approximately 30 km altitude were made with balloon-borne University of Wyoming optical aerosol counters. Measurements usually occurred between 6 and 9am, local time, with measurement frequency varying from monthly to bi-monthly. Data are available from 1971 to present. Over this time period three different primary instrument types were used. The latest style (Laser particle counters, LPCs) of the three instrument types was used initially in 2006, became the standard Laramie instrument in 2008, and was, as an example, also flown on quasi-Lagrangian balloons in Antarctica in 2010 (Ward et al., 2014). While the transition from the first instrument to the second was documented in Deshler et al. (2003), a similar study to compare the third Wyoming instrument with the second instrument is a work in progress. For the MIPAS validation, measurements from the second and third Wyoming instruments were available. The positive bias of MIPAS aerosol volumes from the in situ measurements was generally consistent between MIPAS and both of the Wyoming instruments above 20 km. Below 20 km the in situ measurements diverged from each other, with the second instrument indicating higher volumes than the LPC (third instrument), and at times higher than MIPAS. Based on these comparisons with both instruments the Wyoming measurements to be used were confined to those made with the LPC because it permitted the simplest altitude dependent de-biasing function for the MIPAS aerosol volume densities. The LPC measures particles with radii > 0.08–4.2 µm in eight size classes. ~~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. In our study we use the volume density profiles that are derived from the fitted lognormal size distributions (unimodal or bimodal) to the~~

~~measurements. The precision of these volume estimates is ± 40 % (Deshler et al., 2003).~~ Deriving geophysical quantities from the size resolved aerosol concentration measurements requires fitting a size distribution to the in situ data. In the past this has been done by fitting either a unimodal or bimodal lognormal size distribution to a subset of the measurements. The final size distribution parameters selected are those from that subset of the measurements which minimises the root mean square error when the fitted distribution is compared to all the measurements. This approach is transitioning to a new approach which modifies the nominal in situ aerosol sizes based on laboratory measurements of the aerosol counting efficiency. The counting efficiency at each size is then included in a search of the lognormal parameter space for the lognormal coefficients which minimise the error of the fitted distribution, coupled with the counting efficiency, compared to the measurements. In our study we use the volume density profiles that are derived from the fitted lognormal size distributions (unimodal or bimodal, following the new retrieval approach) to the measurements. The precision of these volume estimates is the same as the old method, ±40 % (Deshler et al., 2003). The change in the way the fitting parameters are derived is the subject of a paper to be submitted soon. The impact on size distributions from the LPC measurements is not large.

2.3 Chemical Transport Model

The ~~isentropic~~ Chemical Transport Model (CTM) used in our study (e.g. Sinnhuber et al., 2003; Kiesewetter et al., 2010) is forced by temperature, wind fields, and diabatic heating rates from the ERA-Interim reanalysis (Dee et al., 2011). The model uses isentropes as vertical coordinates. Horizontal transport on levels of constant potential temperature is derived from the wind fields, while vertical transport is calculated using the diabatic heating rates. The CTM employs the second order moments advection scheme by Prather (1986). The model domain covers 29 isentropic levels between 330 and 2700 K (~10–55 km), with a horizontal resolution of about 2.5° latitude x 3.75° longitude (Gaussian latitude grid).

A simple sulfur chemistry scheme ~~is~~ has been implemented during this work, including OCS, SO_2 and H_2SO_4 as advected tracers. In the sulfur scheme no distinction between tropospheric and stratospheric air is implemented. ~~The H_2SO_4 tracer consists of gas-phase and liquid-phase H_2SO_4.~~ In the presented simulations, we consider volcanic SO_2 from one volcanic eruption as the only sulfur source per simulation, in order to study the SO_2 and sulfate aerosol after the eruptions of Kasatochi in 2008 and Sarychev in 2009 individually. Therefore, OCS is not considered as sulfur source in the simulations presented in this work. The bottom boundary concentrations and initial fields are zero for SO_2 and H_2SO_4. The volcanic SO_2 is injected instantaneously into the model column of the volcano. ~~Different~~ SO_2 masses are injected into three altitude ranges (see Sect. 4.1, Table 1), consistent with MIPAS SO_2 observations, and SO_2 is uniformly distributed to the ~~grid boxes~~ air mass per altitude range. The sulfur released from volcanic SO_2 reacts with OH (hydroxyl radical) to form H_2SO_4 ~~by its reaction with OH builds H_2SO_4~~. This is the only chemical reaction considered in the simulations presented in this study. As the interim product sulfur trioxide (SO_3), formed during the oxidation of SO_2, combines rapidly with water vapour to form sulfuric acid, it is not explicitly considered in the model. The sinks for the species are advective transport out of the model domain into the troposphere, removal of SO_2 by its reaction with OH, and gravitational settling for sulfate aerosol. An OH climatology of daily distributions was derived in a full chemistry run of the CTM (2003–2006), and for reaction rates the recommendations from JPL (Jet Propulsion Laboratory) are used (Burkholder et al., 2015). For the sedimentation of sulfate aerosol~~,~~ equilibrium partitioning between gas- and liquid-phase is assumed for H_2SO_4 (Ayers et al., 1980) at each simulation time step. To determine the terminal velocity for the part of the sulfate aerosol that settles, velocity calculations follow Jacobson (1999). In the simulations presented here the ~~sedimentation~~ aerosol radius is fixed to one effective settling radius. The solution density of the aerosol is calculated online from the fraction of liquid-phase H_2SO_4 in the binary solution of the H_2SO_4–H_2O aerosol. The sulfur scheme hydrates the sulfate aerosol based on ambient water vapour loading ~~content~~, which is ~~fixed at~~ assumed to be 4.5 ppmv~~ for the stratosphere~~. Sedimentation transports sulfate aerosol into the grid box below, or finally out of the model domain. All model results shown for H_2SO_4 only consist of the sulfate aerosol droplets, as the MIPAS measurements do not consider gas-phase H_2SO_4.

The model is run for 365 d per simulation, with a time step of 30 min and tracer fields are written out daily at 12 UTC. For the eruption of Kasatochi (7 Aug 2008) the individual runs are started on the 31 Jul 2008, and for the eruption of Sarychev (12 Jun 2009) all runs are started on the 31 May 2009. As the initial tracer fields are set to zero and the model is driven by ERA-Interim reanalysis data, which are updated every six hours, no long spin up time is needed. Per volcano four simulations were made that differ concerning the particle size of sulfate aerosol. Simulations were made with constant radii of 0.1, 0.5 and 1 µm, and without sedimentation. In the atmosphere the radius of sulfate aerosol varies (Deshler et al., 2003 and 2008). Nevertheless, for simplification we use a constant “effective sedimentation radius” to determine the terminal fall velocity, which we consider to be the average settling speed of aerosol particles of different radii. In our simple sulfur scheme, no scavenging of SO_2 or H_2SO_4 by clouds is considered in the model. This would be confined mostly to tropospheric altitudes and in our study region ($\geq$ 10 km) especially to tropical latitudes. Washout by precipitation might play a role there but it is expected to have a minor effect on our study, as we analyse the sulfur that remains in the atmosphere (above ~10 km) after the first weeks following the volcanic eruptions. Furthermore, no nucleation or growth processes of sulfate aerosol are considered.

3 The new MIPAS aerosol ~~dataset~~ data set

3.1 Aerosol retrieval from MIPAS limb-spectra

In previous analyses of mid-infrared observations by ~~MIPAS/Balloon~~ MIPAS-B (the balloon-borne predecessor of the MIPAS satellite instrument; Friedl-Vallon et al., 2004) and MIPAS/Envisat (MIPAS instrument on the satellite Envisat, generally referred to as “MIPAS” throughout the present work) it has been demonstrated that the limb radiance due to particles has two major contributing terms: (1) the thermal emission of the particles, and (2) the scattered radiation from the atmosphere and Earth’s surface from below the tangent point (Höpfner et al., 2002 and 2006). The relative weights of these contributions differ with particle size and wavenumber. For particles sufficiently small compared to the wavelength (d <

~1 µm in the mid-IR; Höpfner, 2004), the scattered contribution can be neglected such that only the thermal emission remains as major source of IR radiation. In this regime the radiance only depends on the total aerosol volume density. Typical sizes of the stratospheric aerosol layer particles are less than 1 µm in case of background and enhanced conditions due to medium sized volcanic eruptions (e.g. Deshler et al., 2003). Thus, our retrieval target is the altitude profile of volume densities, derived from each set of calibrated MIPAS limb-scan spectra.

For this study we have concentrated on the second MIPAS measurement period between January 2005 and April 2012. The retrieval model used is the KOPRA/KOPRAFIT (Karlsruhe Optimized and Precise Radiative transfer Algorithm) suite, allowing to directly retrieve aerosol parameters from observed radiances by coupling a Mie-model with the line-by-line radiative transfer scheme (Stiller et al., 2002; Höpfner et al., 2002 and 2006). For aerosol composition we assume a 75 percent by weight (75 wt%) H_2SO_4–H_2O solution, as the stratospheric sulfuric aerosol composition typically varies between around 70 and 80 %, as obtained by equilibrium calculations (Carslaw et al., 1995) and observations (e.g. Doeringer et al., 2012). ~~E.g.~~ Kleinschmitt et al. (2017), calculating aerosol optical properties, Kremser et al. (2016), calculating sulfur fluxes, and Gao et al. (2007), calculating atmospheric volcanic aerosol loadings, also use a 75 wt% H_2SO_4–H_2O composition in their studies. The imaginary parts of various refractive index ~~datasets~~ data sets in the mid-IR are displayed in Fig. 1. Here the used optical constants by Niedziela et al. (1999) for 75 wt% and 230 K (bold red line) are compared to data at other concentrations and temperatures by Niedziela et al. (1999) (upper panel), and Myhre et al. (2003) (lower panel). This particular data set has been chosen because in an evaluation of optical constants for sulfuric acid, Wagner et al. (2003) have found those ~~datasets~~ data sets to be best consistent with observations in the aerosol chamber AIDA (Aerosol Interactions and Dynamics in the Atmosphere). The spectral range selected for the retrieval (1,216.5–1,219.5 cm^{-1}) is situated at the long wavelength end of MIPAS band B as indicated by the two vertical lines in Fig. 1. It lies within one of the atmospheric windows as can be seen by comparison to the limb-transmission curve (light grey) in Fig. 1. We have not chosen the windows at around 830 cm^{-1} and 950 cm^{-1} since at 1,220 cm^{-1} the absorption by H_2SO_4 droplets is higher and the relative difference between the various sets of refractive indices is ~~lower~~ smaller.

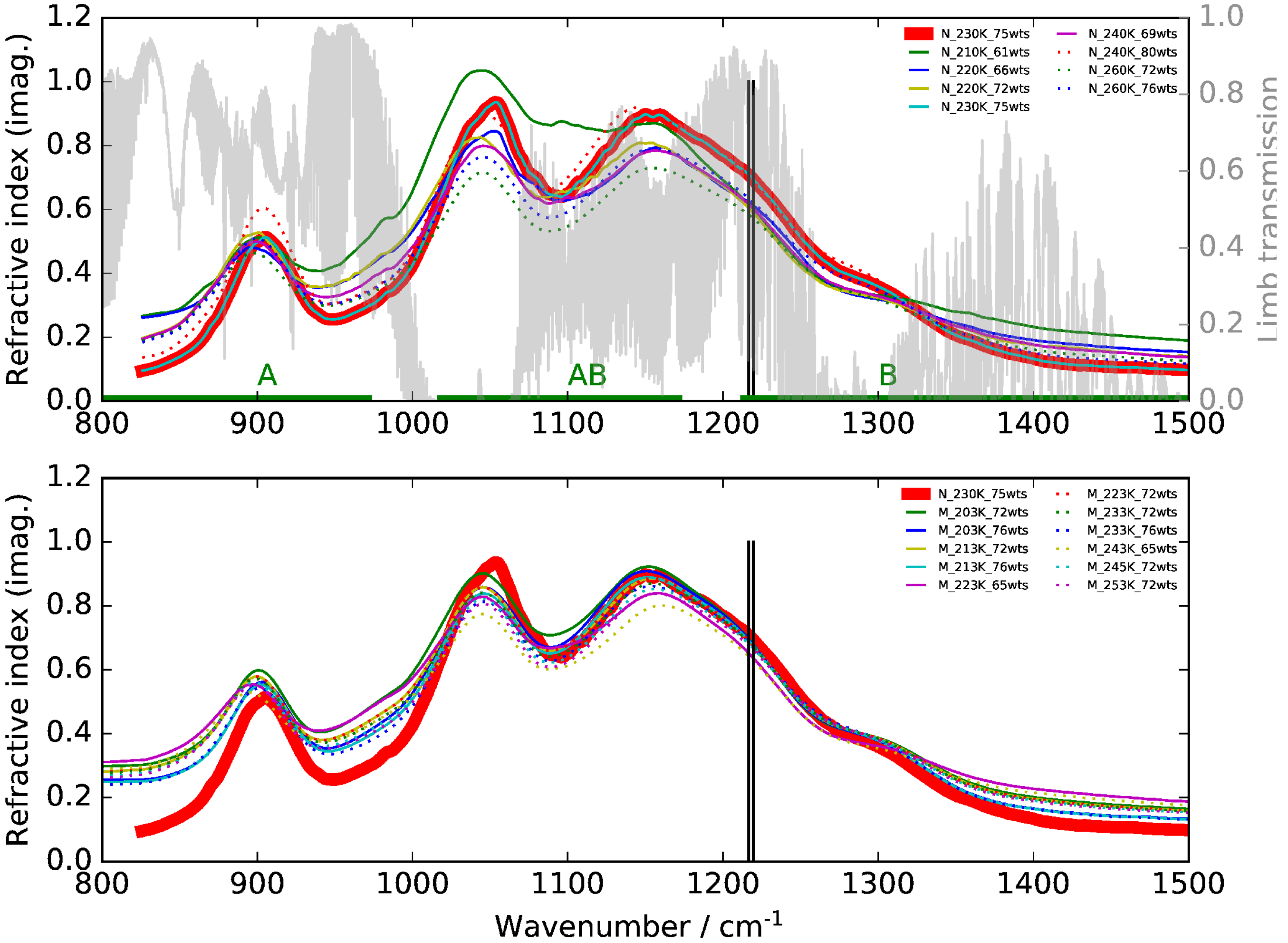

Figure 1. Imaginary parts of refractive indices for aqueous H_2SO_4 solutions of different concentrations (wts: weight % H_2SO_4) and different temperatures in the mid-infrared region. **(Top)** thin solid and dotted curves: Niedziela et al (1999), **(bottom)** Myhre et al. (2003). The bold red line indicates the ~~dataset~~ data set, and the two vertical lines the spectral window used in this study. A simulated limb transmission spectrum for 10 km tangent altitude for standard mid-latitude conditions is additionally plotted in the top row as well as the wavenumbers covered by MIPAS bands A, AB and B.

The retrieval has been set up as a multiparameter nonlinear least-squares fit of the calculated to the measured limb radiances of entire limb-scans (e.g. von Clarmann et al., 2003). Besides the target parameter, namely sulfate aerosol volume densities, further atmospheric fit-parameters of the retrieval are vertical profiles of spectrally interfering trace gases methane (CH_4), H_2O, ozone (O_3), and nitric acid (HNO_3). While zero initial guess profiles have been used for the volume densities, results from the IMK routine processing are taken for the trace gases (von Clarmann et al., 2009). As the atmospheric parameters are represented at denser altitude levels (1 km) than the vertical field-of-view (~3 km) and the vertical tangent point spacing (1.5 km) of MIPAS, constraints on the smoothness of the profile shape are introduced by ~~regularization~~

regularisation (Tikhonov, 1963; Steck, 2002). The retrieval of aerosol volume density is restricted to altitudes up to 33 km and the ~~regularization~~ regularisation strength has been adjusted such that its resulting vertical resolution is around 3 to 4 km. To cover instrumental uncertainties a spectral shift parameter and a radiance offset, constant over all wavenumbers and tangent altitudes, has been retrieved simultaneously to the atmospheric quantities. For the analysis in this paper only data at altitudes with averaging kernel diagonal elements larger than 0.05, which are at least 1 km above the lowest tangent height, are used.

An overview of the leading error components is presented in Fig. 2, with the assumed parameter uncertainties listed in the caption. The error contributions are estimated from a subset of a few hundred single cases by sensitivity studies using modified parameters or, in case of spectral noise, directly from the retrieval diagnostics. The total error changes with altitude from around 20 % (0.09 μm^3 cm^{-3}) at 10 km up to over 40 % (0.005 μm^3 cm^{-3}) at 30 km. It is dominated by the uncertainty of the optical constants resulting in 10–20 % error, followed by tangent pointing knowledge with 5–15 %. The error component resulting from spectral noise is rather constant with altitude in absolute terms of volume density and amounts to about 0.01 μm^3 cm^{-3}. Other instrumental errors that have been investigated but are not listed in Fig. 2 are uncertainties due to the knowledge of the instrumental line shape, and radiometric gain and offset calibration error. In the estimation of the radiative error no offset variations with tangent altitudes were considered, and, thus, compensated for by the retrieval approach. However, a tangent altitude dependent radiometric offset error caused e.g. by straylight in the instrument cannot be excluded (López-Puertas et al., 2009). We have not handled this uncertainty in the framework of error estimation but we have tried instead to compensate for it through a de-biasing of the ~~dataset~~ data set based on validation with in situ observations as described in Sect. 3.2.

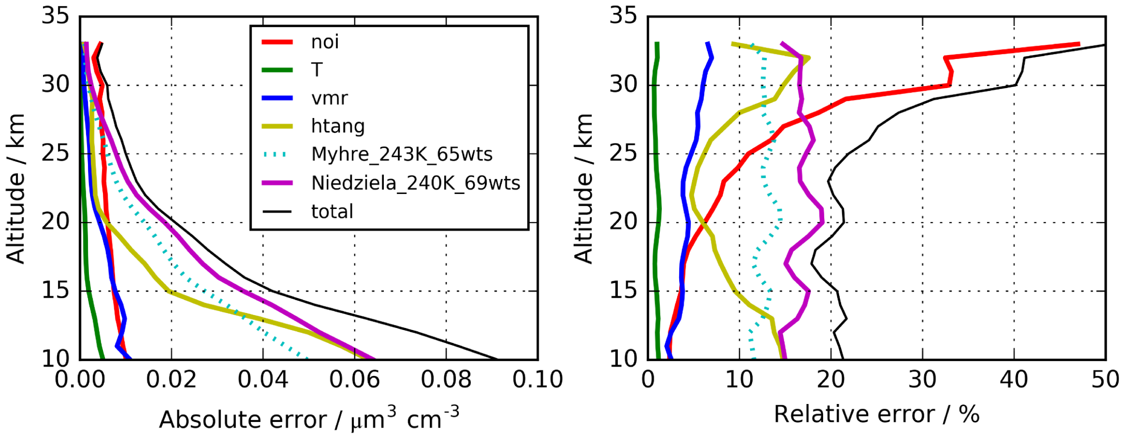

Figure 2. Altitude dependent estimated errors for the retrieval of H_2SO_4–H_2O aerosol volume densities. Solid lines indicate the uncertainties used to calculate the 'total' error. Indicated errors are: 'noi': single scan spectral noise, 'T': temperature uncertainty 2 K, 'vmr': 10 % uncertainty of volume mixing ratios of interfering gases, 'htang': 300 m tangent altitude uncertainty, 'Niedziela_240K_60wts': use of optical constants by Niedziela et al. (1999) for 240 K and 69 wt% instead of Niedziela et al. (1999) for 230 K and 75 wt% H_2SO_4. The dotted curves ('Myhre_243K_65wts') show the results when the optical constants of Myhre et al. (2003) for a temperature of 243 K and a concentration of 65 wt% are used instead of those by Niedziela et al. (1999) (T=230 K and 75 wt%).

Prior to the retrieval, a deselection of spectra affected by clouds has been performed via application of an established cloud filter method for MIPAS by Spang et al. (2004). To sort out optically thick clouds, but not all aerosol-affected spectra, this cloud filter has been applied with a cloud index limit of 1.7. Due to this loose setting of the cloud-filter artefacts caused e.g. by thin cirrus clouds, polar stratospheric clouds (PSCs) or volcanic ash remain in the ~~dataset~~ data set, which are all attributed to the retrieved 75 wt% H_2SO_4–H_2O aerosol volume density. Thus, further filtering of affected profiles has been necessary after completing the retrieval.

Two distinct features of strong enhancements with an annual cycle show up in the unfiltered ~~dataset~~ data set. The first feature is due to strong enhancements in the presence of PSCs at the winter pole. To deselect PSC-affected profiles a filter is applied when temperatures in the altitude range 17–23 km drop below a threshold of 195 K polewards of 40°, for the Northern Hemisphere from 15 Nov–15 Apr, and for the Southern Hemisphere from 1 Apr–30 Nov. This temperature represents the nitric acid trihydrate (NAT) existence temperature at around 20 km~~,~~ under typical stratospheric conditions for nitric acid~~ (HNO_3)~~ and water vapour~~ (H_2O)~~.

The second feature is assumed to be induced by thin cirrus clouds. It is present mainly in the tropics, at around 25° S–25° N, at altitudes between about 13–21 km. It reaches highest altitudes and is most intense above and in the vicinity of continents, and above the western Pacific. The vertical extent is smallest in boreal summer, and its vertical gradient towards lower aerosol volume densities is relatively strong, with no upward transport being observed. Both~~,~~ the location in the tropics, in regions of strong vertical motions and convective clouds, and the relatively sharp decrease at higher altitudes towards increasing temperatures, suggest that it is connected to the influence of ice particles. The ice-filter for MIPAS data by Griessbach et al. (2016) is applied on all retrieved MIPAS aerosol profiles to reduce the effect of spectra influenced by ice in the present ~~dataset~~ data set. ~~This~~ Their method consists of two steps to detect whether MIPAS spectra are influenced by aerosols, ice, clouds, ashes, or a clear sky (Griessbach et al., 2014 and 2016). First aerosols and clouds are identified, using a spectral window region that is sensitive to aerosols and clouds. Then ice clouds and aerosols are discriminated, using spectral windows with contrasting behaviour for ice and aerosols. This is then combined to brightness temperature difference correlations. In our ~~dataset~~ data set, we consider only retrieved values starting 4 km above the altitude of the uppermost spectrum that was flagged to have been influenced by ice. Further, the ash filter for MIPAS spectra by Griessbach et al. (2014), based on an ash detection threshold function, is applied in the same way as the ice filter, to filter out volcanic ash and mineral dust.

3.2 Validation and bias correction

To validate the new ~~dataset~~ data set, we compare the profiles of MIPAS aerosol volume density to in situ balloon measurements (Deshler et al., 2003). In situ measurements were carried out with laser based aerosol spectrometers ~~(LPCs)~~ from Laramie, Wyoming (41° N/105° W), between 6 and 9am, local time. In Fig. 3, profiles of the balloon measurements are shown. In comparison, MIPAS mean aerosol volume density profiles are presented, selected from a restricted area around Laramie, together with their normalised standard deviations (Fig. 3), which show the variability of the underlying profiles. MIPAS profiles are chosen for the day of the balloon flight~~,~~ within a 5° latitude x 10° longitude distance around Laramie. Further, in Fig. 4a we show the mean over ~~all~~ the profiles that were retrieved from LPC measurements, as shown in Fig. 3 (excluding the 28.07.2011) and the corresponding MIPAS profiles, together with the absolute (b) and relative difference (c) of the mean MIPAS profile to the mean profile from balloon measurements. In Fig. 4a the standard errors of the mean ~~show the uncertainty of the bias~~ profiles are presented, and in Fig. 4b the statistical uncertainty of the bias (difference between the mean in situ and mean MIPAS profile) is shown. The profile on the 28.07.2011 shows large differences between MIPAS and the in situ data and the strongest vertical variability of all in situ profiles at low altitudes (below ~18 km), possibly due to the Nabro eruption (12 Jun 2011). Hence it is excluded from the calculation of the mean profiles shown in Fig. 4.

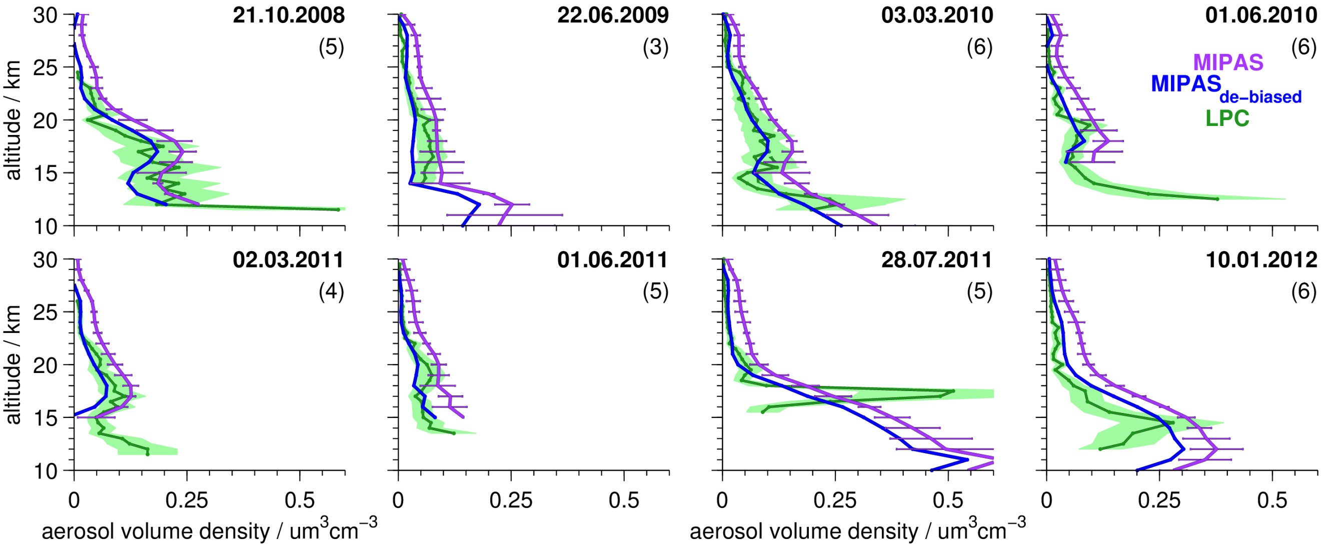

Figure 3. Profiles of aerosol volume densities from in situ and MIPAS satellite measurements. In situ data are balloon-borne data above the lapse rate tropopause for Laramie, Wyoming (41° N/105° W), measured by Laser Particle Counters ~~(LPCs)~~. Two instruments were flown on the 03.03.2010. The 40 % errors of the in situ data are shown as green shaded areas. MIPAS satellite data in purple (original data) and blue (de-biased data). For all MIPAS data the ice filter by Griessbach et al. (2016) and ash filter by Griessbach et al. (2014) , and our PSC filter are applied. MIPAS profiles are averages of all available profiles for the indicated day, averaged in a ± 5° lat x 10° lon area. In brackets are the number of MIPAS profiles included in the average. For the non-de-biased MIPAS values the standard deviations of the mean profiles are shown.

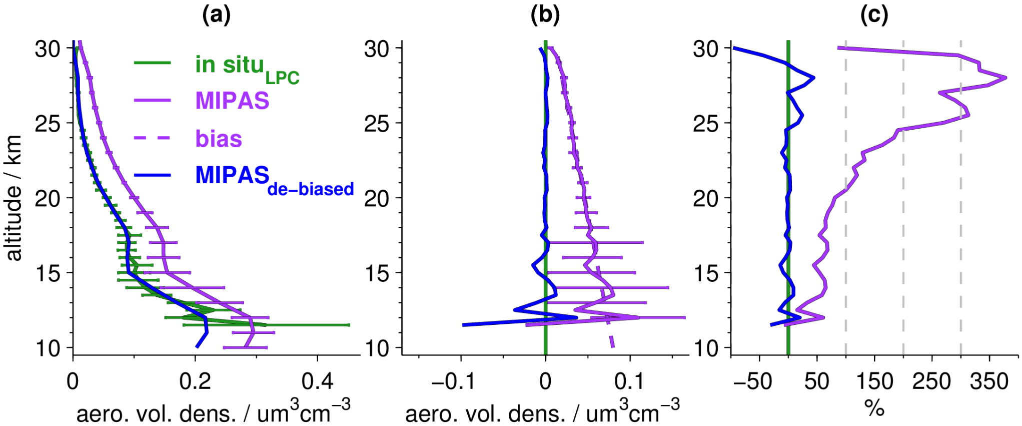

Figure 4. Aerosol volume densities by MIPAS and in situ data from Laramie, Wyoming. **(a)** mean profiles for data in Fig. 3 (excluding the 28.07.2011), and their 1-sigma standard errors. In blue are de-biased MIPAS values, in purple the original data. **(b)** absolute and **(c)** relative differences of MIPAS to LPC in situ data. Differences are calculated for the mean profiles shown in (a). In (b) the statistical uncertainty of the absolute difference between in situ and MIPAS data is shown (horizontal purple lines; square root of the sum of the 1-sigma standard errors squared for MIPAS and the in situ data). Dashed purple line: linear fit to the original MIPAS data between 18 and 30 km (purple solid line in panel b), used to reduce the bias towards the LPC in situ measurements.

Generally, the aerosol volume densities (Fig. 3 and 4) are highest in the lower stratosphere and then decrease towards zero at higher altitudes. As the balloon data ~~has~~ have a higher vertical resolution, and the retrieval process for MIPAS profiles includes smoothing, the in situ data show finer structures. Compared to the balloon data, the original MIPAS aerosol volume densities show a positive bias in most profiles (Fig. 3) as well as in the mean profile (Fig. 4). This is most easily detectable at higher altitudes where profiles are relatively smooth. The offset amplifies towards lower altitudes (Fig. 4b). Aiming on a reduction of this positive offset, a height-dependent de-biasing is performed on all single MIPAS profiles. The de-biasing is based on the in situ measurements carried out with laser based particle counters. MIPAS profiles show a consistent variation with height, compared to the LPC measurements. An additive linear de-biasing is applied, rather than a multiplicative correction, as the offset is expected to be caused by ~~a possible~~ an altitude-dependent additive stray light error in the radiances (see Sect. 3.1). ~~The offset-correction is represented in Fig. 4b (dashed line). It is the difference between a linear fit to MIPAS values at 18 to 30 km altitude, and the in situ data (Fig. 4b, pink and blue solid line, respectively).~~ The de-biasing is based on the absolute differences between the aerosol volume densities of the mean MIPAS and in situ profiles (Fig. 4b, purple solid profile) at 18–30 km where profiles show weak variability and low uncertainty of the bias. A linear least squares fit (Fig. 4b, purple dashed line) to the profile of absolute differences represents the vertically resolved values of the de-biasing, which are subtracted from each MIPAS profile during offset-correction. At lower altitudes~~,~~ where profiles show more variability, both vertically and between the in situ and MIPAS profile, this linear fit also suits well. The uncertainty of the bias (Fig. 4b) at altitudes above ~17 km shows that the positive bias is not random, as the spread is low and uncertainty limits are noticeably distant from zero. The mean de-biased MIPAS profile (Fig. 4a) matches the in situ data and lies mostly in the range of the standard error of the mean of the in situ data. Further, the absolute and relative differences to the balloon data are reduced significantly (Fig. 4b and c). Percentage differences are mostly below ± 25 %. For the non-de-biased profile, at altitudes above around 20 km, percentage differences increase strongly~~,~~ due to very low aerosol volume densities, while at lower altitudes percentage differences are below about 100 %. By excluding the in situ and MIPAS profiles measured on 28.07.2011 in the calculation of the mean profiles, the agreement between the measurements is improved in the altitude range below 18~km, while above this altitude changes are marginal, as can be expected from Fig.~3. The de-biasing is therefore not affected by the dismissal of the observations from this day.

3.3 Time series of MIPAS sulfate aerosol and SO_2 for 2005 to 2012

To study the distribution of sulfate aerosol~~,~~ as measured by MIPAS from 2005 to 2012, Fig. 5 (left) shows latitudinally resolved time series of liquid-phase H_2SO_4 mole-fractions, for various altitudes from 10 to 22 km. From the retrieved aerosol volume densities, the mole-fractions are calculated by assuming all ~~the~~ aerosol to be sulfate aerosol with a composition of 75 wt% H_2SO_4 and 25 wt% H_2O, and an aerosol density of 1,700 kg m^{-3}. Strongest variability in the MIPAS sulfate aerosol data is caused by volcanic eruptions. In the Northern Hemisphere, strongest signatures of volcanic eruptions are due to the eruptions of Kasatochi (52.2° N/175.5° W) in August 2008, Sarychev (48.1° N/153.2° E) in June 2009, and Nabro (13.4° N/41.7° E) in June 2011, at altitudes from 10 to about 20 km. At low latitudes~~,~~ tropical volcanoes such as Manam (4.1° S/145.0° E) in January 2005, Soufrière Hills (16.7° N/62.2° W) in May 2006, and Rabaul (4.3° S/152.2° E) in October 2006, increase the sulfate aerosol mole-fractions at higher altitudes, above 16 km. The aerosol is lifted upwards with time and the plumes get modulated by the Quasi-Biennial Oscillation in the tropics. A similar pattern of upward motion of the volcanic aerosol from these tropical eruptions has been seen in satellite measurements of aerosol extinction ratios (Vernier et al., 2011). In the Southern Hemisphere the eruption of Puyehue-Cordón Caulle (40.6° S/72.1° W) in June 2011 has the strongest impact on the measurements, but is restricted to lower altitudes, below about 14/15 km. During the preceding years the mole-fractions are relatively low in the mid-latitudes of the Southern Hemisphere, at 10 to 12 km.

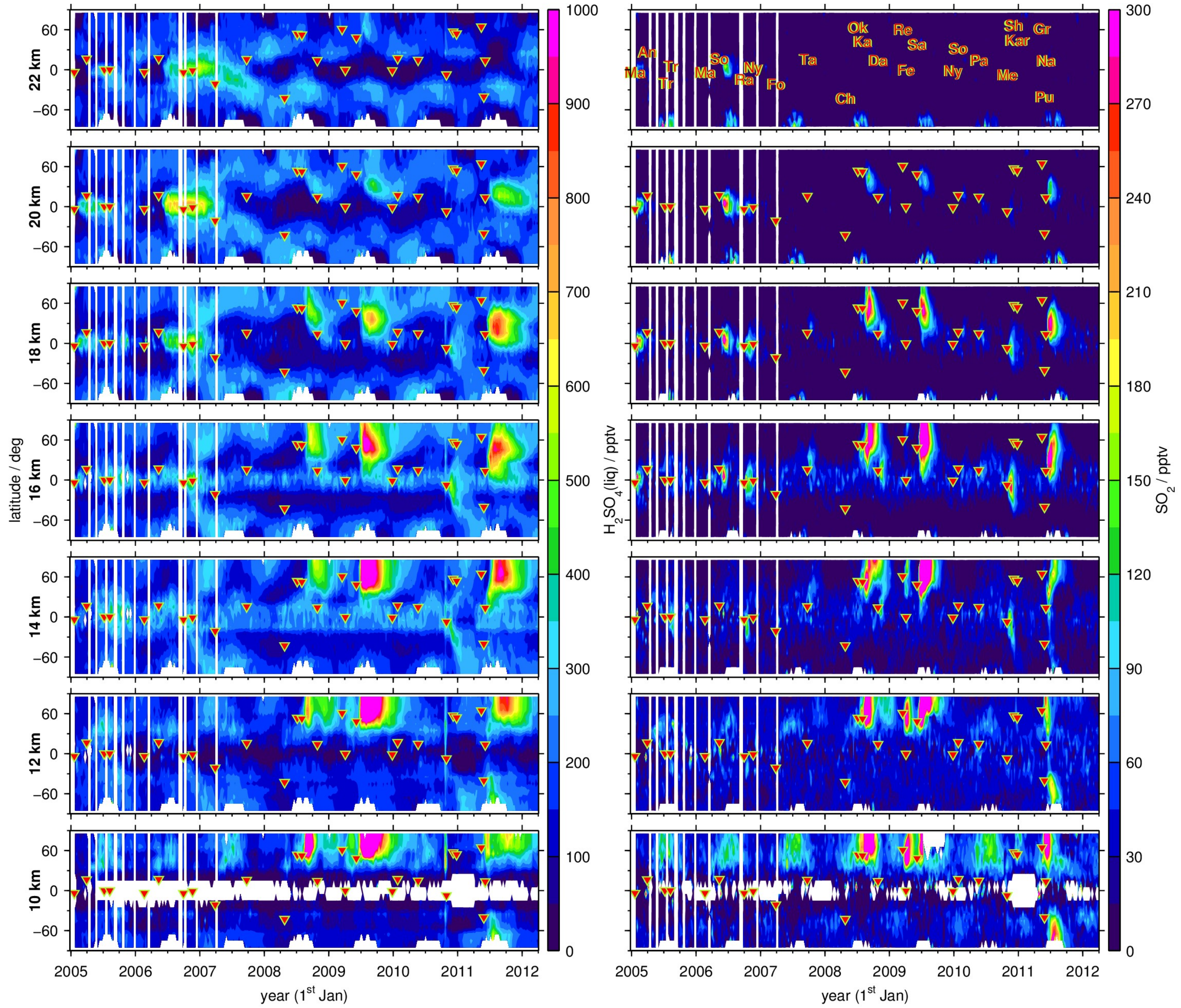

Figure 5. Global time series of latitudinally resolved distributions of MIPAS liquid-phase H_2SO_4 **(left panels)** and SO_2 **(right panels)** volume mixing ratios. Shown are 8-day mean values for 10° latitude bins at different altitudes (10 to 22 km). The colour-code is restricted to 0–1,000 pptv for H_2SO_4(liq), and 0–300 pptv for SO_2. Values exceeding these limits are assigned with the limiting value, respectively. Volcanic eruptions are indicated by red triangles (together with their abbreviated names in the uppermost right panel; following ~~from~~ Höpfner et al., 2015~~; Table 3 therein~~). Abbreviations: An: Anatahan, Ch: Chaiten, Da: Dalaffilla, Fe: Fernandina, Fo: Piton de la Fournaise, Gr: Grímsvötn, Ka: Kasatochi, Kar: Karymsky, Ma: Manam, Me: Merapi, Na: Nabro, Ny: Nyamuragira, Ok: Okmok, Pa: Pacaya, Pu: Puyehue-Cordón Caulle, Ra: Rabaul, Re: Redoubt, Sa: Sarychev, Sh: Shiveluch, So: Soufrière Hills, Ta: Jebel at Tair, Tr: unidentified Tropical Volcano.

The latitudinally resolved time series of sulfate aerosol mole-fractions further reveal different periodic structures, which are not connected to volcanic activities:

1. In polar regions at altitudes above ~16 km, sulfate aerosol mole-fractions decrease strongly in winter to spring. The pattern is more pronounced in the Southern Hemisphere. This decrease is connected to the polar vortex, where relatively sulfate aerosol free air is transported downwards. Thomason and Poole (1993) reported on very low observed aerosol levels relative to non-vortex air.
2. In both hemispheres, but primarily in the Southern Hemisphere, mole-fractions of liquid-phase H_2SO_4 are enhanced at around 20–22 km in the mid-latitudes (and partly the tropics) during boreal / austral winter and spring, respectively. In the stratosphere sulfur ~~Sulfur~~ is released from OCS mainly in the tropics at altitudes between about 25 and 35 km (Brühl et al., 2012) and the sulfate aerosol that is produced ~~built~~ is transported towards mid-latitudes and lower altitudes.
3. In the mid-latitudes of the Northern Hemisphere, the sulfate aerosol is increased during boreal summer at around 10–12 km.
4. In the tropics at around 14–16 km~~,~~ aerosol values are elevated, while they are very low below and above these altitudes, unless influenced by volcanic eruptions.

As SO_2 is the main precursor for stratospheric sulfate aerosol during volcanically perturbed times, we analyse similarities and discrepancies between the distribution of latitudinally resolved time series of MIPAS SO_2 at various altitudes for 2005–2012 (Fig. 5, right), and the new aerosol data (Fig. 5, left). Prominent features seen in the distribution of SO_2 mole-fractions from single limb scans are described by Höpfner et al. (2015). It should be noted, that these data are useful mainly for the analysis of enhanced SO_2, rather than for background conditions, for which the monthly and zonal mean MIPAS SO_2 ~~dataset~~ data set by Höpfner et al. (2013) is more suited. The distributions of SO_2 and sulfate aerosol show clear similarities, especially concerning volcanic plumes. Differences in the patterns result mostly from the longer residence time of sulfate aerosol in the stratosphere, compared to SO_2. Sulfate aerosol can reside in the stratosphere for several months up to several years, if it is neither being transported back to the troposphere nor evaporated at higher altitudes. On the contrary, SO_2 has a stratospheric lifetime of a few weeks. Different point sources, such as volcanic eruptions, can therefore be distinguished more easily in the SO_2 measurements than in the aerosol data. Further discrepancies arise from the fact that sulfur is released from SO_2 over weeks, during the exponential decay of the latter, and can then be converted into sulfate aerosol. Elevated SO_2 amounts are therefore not instantly leading to elevated sulfate aerosol amounts, and the curve of enhanced sulfate aerosol is broader and flatter than for SO_2.

In the Northern Hemisphere at low altitudes (< 12/13 km) during boreal summer a similar feature of increased VMRs is present in the SO_2 as in the sulfate aerosol data (point 3). A closer look at the monthly distribution of SO_2 and sulfate aerosol reveals no distinct patterns (for SO_2 see Höpfner et al., 2015); enhancements are spread over the entire Northern Hemisphere. In Höpfner et al. (2015) this feature could not be confirmed due to a lack of SO_2 in situ data. The presence of similar enhancements in the aerosol data supports the hypothesis of the increased sulfur loading ~~content~~ at low altitudes in the Northern Hemisphere not being a retrieval artefact. Further, elevated values in the tropics at around 14–16 km, as seen in the

MIPAS aerosol (point 4) are also present in the SO_2 data. These are ~~localized~~ localised mostly in continental regions, and the western Pacific, both for MIPAS sulfate aerosol and SO_2 (for SO_2 see Höpfner et al., 2015).

4 Volcanic eruptions of Kasatochi in 2008 and Sarychev in 2009

In this section a case study of MIPAS SO_2 and sulfate aerosol measurements and CTM model simulations for the two volcanic eruptions of Kasatochi and Sarychev (Kasatochi: 7 Aug 2008, 52.2° N/175.5° W; Sarychev: 12 Jun 2009, 48.1° N/153.2° E) is presented. Both volcanoes erupted at Northern Hemisphere mid-latitudes during boreal summer. MIPAS satellite measurements are compared to CTM simulations, to study the ~~fate~~ evolution of the emitted sulfur in terms of conversion from SO_2 to sulfate aerosol, and its transport and removal at altitudes between 10 and 22 km. As our intention is to study explicitly the sulfur per volcanic eruption, background values per model simulation are set to zero for both SO_2 and H_2SO_4, and no other sources than the volcanically emitted SO_2 of one volcanic eruption is included.

4.1 Sulfur mass in the Northern Hemisphere mid- and high-latitudes

In this section we aim at testing the agreement between measured SO_2 and liquid-phase H_2SO_4 masses, together with modelled data, in terms of the increase and decline of sulfur emitted by the volcanic eruptions of Kasatochi in August 2008 and Sarychev in June 2009, and the influence of ~~sedimentation~~ the prescribed effective sedimentation radius on the residence time of sulfate aerosol. ~~Good agreement between the modelled and measured SO_2 masses is essential to test, by comparison with modelled sulfate aerosol, if the measured aerosol is quantitatively and qualitatively consistent with its measured precursor.~~ As we intend to test if the measured aerosol is quantitatively and qualitatively consistent with its measured precursor by comparison with modelled sulfate aerosol, a good agreement between the modelled and measured SO_2 masses is essential.

In Table 1~~,~~ injected SO_2 ~~masses~~ amounts for three altitude regions are given (labelled 'present study'), as used for the CTM simulations in the present study, ~~are given (labelled 'present study'),~~ together with comparisons to volcanic SO_2 masses from the literature. ~~The simulations result in good agreement between measured and modelled SO_2.~~ The upper injection limit for the volcanic emissions in the CTM is set to 19 km~~, an altitude limit derived from comparisons with MIPAS SO_2~~. Simulations have been made with varying injected SO_2 masses and upper injection altitude limits, intending to achieve good agreement between the modelled and measured SO_2 masses (comparisons as in Fig. 6). The data presented here resulted in the best agreement, with comparisons starting approximately one month after the respective eruption (explanation in the following). Due to the limited number of simulations no uncertainties are given for the presented SO_2 masses. The main part of SO_2 per eruption is emitted into the altitude region from 10 to 18 km, and only few percent of the SO_2 masses are injected into altitudes above 18 km. Our best match for Kasatochi is consistent with the lower limit of Höpfner et al. (2015).~~In the case of Kasatochi, our model is run with the SO_2 masses by Höpfner et al. (2015), reduced by their given uncertainties.~~ Höpfner et al. (2015) derived volcanic SO_2 masses for three altitude regions from 10–14 km, 14–18 km, and 18–22 km by exponential extrapolation of the MIPAS SO_2 masses back to the eruption day. ~~This method was applied~~ They applied this method as in the first month after the eruption MIPAS underestimates the SO_2 (Höpfner et al., 2015). Their method results in relatively large error bars that depend on the time period the fit is based on (Höpfner et al., 2015; presented also in Table 1). ~~Error bars of the extrapolated values were found to be relatively large (Höpfner et al., 2015; presented also in Table 1). In the case of Sarychev, the SO_2 mass used in the present study lies below the given error bars~~. For Sarychev, however, our best estimate is smaller than the error limits of the SO_2 masses given by Höpfner et al. (2015).

When comparing the SO_2 masses from different studies, it has to be pointed out, that the SO_2 masses are generally not derived for the same altitude regions. Höpfner et al. (2015), Brühl et al. (2015), and the present study are not totally independent from each other, as they are entirely or partly based on the same MIPAS SO_2 data by Höpfner et al. (2015). The SO_2 masses in our study lie below all studies but Brühl et al. (2015) for Kasatochi, and in the range of the other publications for Sarychev. The wide range of SO_2 masses in Table 1 shows the difficulties and uncertainties related to the determination of volcanically emitted SO_2.

Table 1. Volcanically emitted SO_2 masses from various publications for Kasatochi in 2008 and Sarychev in 2009. For Höpfner et al. (2015) the given total uncertainty is the sum of the uncertainties per altitude range (Table 3 therein). In the case of Pumphrey et al. (2015) pressure levels are given. These represent the highest pressure used for vertical integration.

Kasatochi in 2008			**Sarychev in 2009**		
	SO_2 mass Gg	**Height range**		**SO_2 mass Gg**	**Height range**
Present study	**677**	**10–19 km**	**Present study**	**768**	**10–19 km**
	518	**10–14 km**		**401**	**10–14 km**
	124	**14–18 km**		**362**	**14–18 km**
	35	**18–19 km**		**5**	**18–19 km**
Höpfner et al. (2015)	898 (± 221)	10–22 km	Höpfner et al. (2015)	1,474 (± 357)	10–22 km
	645 (± 127)	10–14 km		888 (± 293)	10–14 km
	210 (± 86)	14–18 km		542 (± 60)	14–18 km
	43 (± 8)	18–22 km		44 (± 4)	18–22 km
Brühl et al. (2015)	376		Brühl et al. (2015)	562	
Pumphrey et al. (2015)	1,350 (± 38)	215 hPa	Pumphrey et al. (2015)	~~1,160~~ 571 (± 42)	147 hPa
Clarisse et al. (2012)	1,600			~~571 (± 42)~~ 1,160 (± 180)	215 hPa
Karagulian et al. (2010)	1,700		Clarisse et al. (2012)	900	
Prata et al. (2010)	1,200		Haywood et al. (2010)	1,200 (± 200)	
Kristiansen et al. (2010)	1,000	> 10 km	Carn et al. (2016)	1,200	
Krotkov et al. (2010)	2,200				
Corradini et al. (2010)	900–2,700				
Thomas et al. (2011)	1,700				
Carn et al. (2016)	2,000				

Time series of sulfur mass contained in SO_2 and sulfate aerosol are shown in Fig. 6~~,~~ for observations by MIPAS and simulations by the CTM. The sulfur loading ~~content~~ is shown for three altitude regions, from 10.5–14.5 km, 14.5–18.5 km and 18.5–22.5 km, for 30° N–90° N, including the latitude of the eruptions (~50° N). For liquid-phase H_2SO_4~~,~~ four model results are presented for each volcanic eruption. The simulations differ concerning the implemented sedimentation (no sedimentation and effective sedimentation radii of 0.1, 0.5, and 1 µm)~~effective sedimentation radius (0, 0.1, 0.5, and 1 µm)~~. In Fig. 6a–c, the sulfur mass is shown for SO_2 and sulfate aerosol separately, while Fig. 6d–f ~~is~~ presents the total sulfur contained in SO_2 and sulfate aerosol. The total simulated sulfur mass is not influenced by chemical sulfur removal, but only by removal due to transport by advection and sedimentation. In Fig. 6, the eruption times of Kasatochi (7 Aug 2008) and Sarychev (12 Jun 2009) are indicated. Additionally, the eruption time of Redoubt (23 Mar 2009) is marked, as this eruption produces a signal in the measurements. It is not included in the simulations, however.

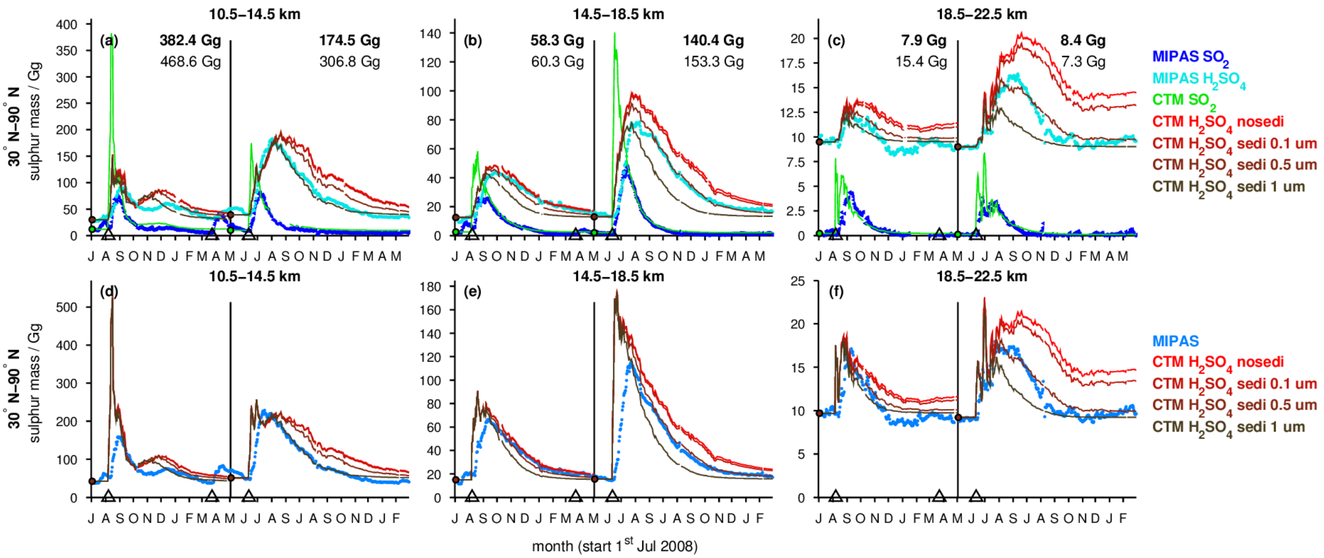

Figure 6. Sulfur mass contained in SO$_2$ and sulfate aerosol after the eruptions of Kasatochi (7 Aug 2008) and Sarychev (12 Jun 2009), from MIPAS measurements and CTM simulations. Presented are 5-days running means for 30° N to 90° N, in three altitude regions (10.5–14.5 km, 14.5–18.5 km, 18.5–22.5 km). For the CTM four simulations for H$_2$SO$_4$(liq) are shown with different effective sedimentation radii (~~0,~~ 0.1, 0.5, and 1 µm) and without sedimentation. All simulations are carried out with the volcanic SO$_2$ masses from Table 1 ('present study'). A constant background is derived from the measured data and added to the simulated sulfur masses per altitude region (indicated by circles). Model simulations for Kasatochi and Sarychev are separated by a vertical black line in May 2009. **(a) to (c)**: sulfur mass per species. **(d) to (f)**: sum of sulfur mass in SO$_2$ and sulfate aerosol. The numbers in (a) to (c) show the peak values of sulfur mass contained in simulated SO$_2$ on locations covered ~~also~~ by MIPAS (bold), and for all available model data. Indicated by black triangles are the eruption days of Kasatochi (7 Aug 2008, 52.2° N/175.5° W), Redoubt (23 Mar 2009, 60.5° N/152.7° W), and Sarychev (12 Jun 2009, 48.1° N/153.2° E).

To analyse the measured and simulated data, ~~datasets~~ data sets of sulfur mass densities (SMD = mass per unit volume) are re-sampled on a common grid with 1 km vertical spacing and a horizontal resolution that equals the model grid. On this new grid, the same data basis is used for the measured and simulated data, dismissing all 'grid cells' for which either only MIPAS or only CTM data are available. For MIPAS aerosols, SMDs are calculated from the primarily retrieved volume densities, using an assumed aerosol density of 1,700 kg m^{-3}, and a binary solution of 75 wt% H$_2$SO$_4$–H$_2$O, while for MIPAS SO$_2$ and the ~~CTM data~~ modelled values, SMDs are calculated from the ~~measured and simulated~~ mole-fractions. Sulfur masses are then derived from 5-days running zonal means of SMDs, by multiplication with the corresponding air volume of the new grid.

Generally, when calculating an integrated mass, high data coverage is crucial to prevent underestimation, therefore we use~~ a~~ 5-days running zonal means. Zonal mean values, used to calculate sulfur masses, are derived using a method of increasing area averaging (see Appendix), to reduce the bias of mean values due to a non-uniform data coverage. Even though high data coverage is very important, we ~~use a method that dismisses~~ dismiss available data and information, as the same basis of available values is used for MIPAS and the CTM. This is appropriate when analysing the agreement between the data. Data ~~is~~ are especially dismissed for the CTM. Thus we also provide some information on modelled sulfur masses derived from the non-co-located data (Fig. 6a–c). The impact of missing data is strongest in the lowermost altitude region presented here. For MIPAS this is mainly due to the presence of clouds and ash, which were filtered out using the cloud filter by Spang et al. (2004) in the case of SO_2 and partly filtered out in the case of aerosol, and additionally the ice and ash filters by Griessbach et al. (2016 and 2014, respectively) for the aerosol retrieval. The CTM has low data coverage at lower altitudes due to its isentropic vertical grid. Interpolation to geometric heights starting at 10 km, produces missing values at altitudes up to 13 km~~, especially in mid- to high-latitudes, and in the presence of strong vortices~~.

To ease visual comparisons of measured and modelled sulfur mass in Fig. 6, a constant background is added to the model results, as only volcanic sulfur is considered in these simulations. The background mass is chosen considering the mass derived by MIPAS before the volcanic eruption in the region of interest, per altitude and latitude bin. This does not necessarily represent normal background conditions, but unmasks the anomalies caused by the volcanoes.

Concerning the measured and modelled SO_2 masses after the eruptions of Kasatochi and Sarychev (Fig. 6), comparisons show that until about one month after the eruptions, the SO_2 mass is by far underestimated by MIPAS. This underestimation of SO_2 was stressed by Höpfner et al. (2015), when comparing MIPAS SO_2 to measurements by the Microwave Limb Sounder (MLS), on board Aura (Pumphrey et al., 2015). It is mainly due to the presence of particles, that hinders MIPAS SO_2 measurements in largely eruption-affected air-parcels and causes a sampling bias towards less volcano-affected air parcels. Through our model simulations we confirm this bias, and the related time scale found by Höpfner et al. (2015). After this first month, the simulated SO_2 agrees well with the measurements by construction.

The decay of SO_2 is well simulated by the CTM, in comparison to the MIPAS measurements. Only oxidation by OH is considered in the model, and we see that the decay of SO_2 can adequately be described by this mechanism. Other processes, as decay by photolysis or reaction with atomic oxygen (O) are not considered, and following the good agreement between measurements and model results, can be neglected at the temporal and spatial scale ~~and region~~ of interest. Inside volcanic plumes chemistry interactions might lead to changes in SO_2-lifetimes (Bekki, 1995). When a high amount of SO_2 gets depleted by hydroxyl radicals, the concentration of the radicals might decrease, which could reduce the speed of further depletion. The good accordance between MIPAS measurements and CTM simulations, which do not account for any feedback on the OH concentrations, suggests~~,~~ that even if such interactions occurred, they did not produce a strong impact on the timescale of months and larger spatial scales.

To investigate the effect of particle sedimentation on the residence time of sulfur after the volcanic eruptions, model simulations with different effective sedimentation radii are performed, ~~including~~ as well as one simulation without any sedimentation. The radii lie in the range of aerosol size distributions~~,~~ as observed by Deshler et al. (2003) and Deshler (2008) for volcanically perturbed periods, and one constant radius is applied for all H_2SO_4 droplets per simulation. Fig. 6 shows the influence of varying the gravitational settling between no settling, and effective sedimentation radii of 0.1, 0.5, and 1 µm.

The amount of sulfate aerosol removed by sedimentation increases with growing particle size, while the time needed for the removal increases for smaller effective sedimentation radii. The sulfur mass contained in liquid-phase H_2SO_4 from a simulation with ~~a particle~~ an effective settling radius of 0.1 µm differs little from a simulation without sedimentation, while effective settling radii of 0.5, and 1 µm show an increasing impact. In the middle and uppermost altitude region, the best agreement between simulated and measured aerosols is found for ~~a radius~~ an effective sedimentation radius of 0.5 µm, for both eruptions. At 10.5–14.5 km, especially in the case of Sarychev, the simulations show temporal disagreement to the decrease of measured aerosol. In the lowermost altitude range sparse data coverage has to be kept in mind, both for the measurements and model results. ~~Here,~~ At these altitudes sulfate aerosol simulated with a radius of 1 µm compares better. A larger effective sedimentation radius seems more appropriate at lower altitudes, as heavier particles can settle faster, and can be removed more rapidly than smaller and lighter particles. These can float in the atmosphere or undergo ascent. The particle size distributions of aerosols can further show natural variation for different volcanic eruptions; therefore some differences in the agreement between modelled and measured data when studying different volcanic eruptions can be expected. Model simulations show that compared to 10.5–18.5 km and compared to small particles, the bigger particles level out faster in the uppermost altitude range studied here. Reasons for this faster removal of the volcanic aerosol are that only little aerosol is injected in the altitude region 18.5–22.5 km, that bigger particles settle faster, and that settling velocities rise with increasing altitude due to the corresponding decrease in air density. In general, we conclude that ~~a particle~~ an effective settling radius of 0.5 µm gives a satisfactory fit between the measurements and simulations for the purpose of studying sulfur mass and sulfur transport in the Northern Hemisphere. Hence, we base all following model results on the CTM runs with ~~a~~ an effective sedimentation radius of 0.5 µm.

We conclude from the comparisons between measured and simulated SO_2 and sulfate aerosol, that the amplitude of the peak of liquid-phase H_2SO_4 and its removal from the studied altitude regions, as measured by MIPAS, is consistent with the measured SO_2, both qualitatively and quantitatively. In the model~~,~~ sulfur is released from SO_2~~,~~ due to its reaction with OH, and sulfate aerosol is consequently ~~built~~ formed. Modelled SO_2~~,~~ that fits well to MIPAS SO_2 measurements in terms of amplitude and decay, releases sulfur and builds H_2SO_4 that in turn matches well to MIPAS sulfate aerosol in terms of amplitude and decrease.

Further, we find that the dominating process on the ~~fate~~ evolution of volcanic sulfur is transport by the Brewer–Dobson circulation out of the region of interest. This becomes obvious when comparing the long-term removal of total modelled sulfur with and without sedimentation to the observed sulfur mass (Fig. 6d–f). In the case of the CTM this excludes all influence by chemical reactions on the removal of volcanic sulfur. Even though consideration of sedimentation of sulfate aerosol with ~~a~~ an effective sedimentation radius between about 0.5 and 1 µm further improves the agreement between model results and observations in 10–22 km altitude, the decay of modelled sulfur mass without sedimentation already compares rather well with the measured decay of sulfur mass. Hereby we see that the removal is dominated by advection rather than sedimentation.

A peak can be seen in the measured and modelled sulfur dioxide and sulfuric acid masses in November / December 2008 (Fig. 6) in the lowermost altitude region (10.5–14.5 km). This peak is caused by downward transport of sulfur in the extra-tropics that has been emitted by the eruption of Kasatochi. In the following section (Sect. 4.2) more details are given on this transport pattern.

In the altitude region of interest, from around 10 to 22 km height, supplementary processes, as the photolysis of gas-phase H_2SO_4, that is important at altitudes above 30 km (Vaida et al., 2003; Brühl et al., 2015), or a meteoritic dust sink (Brühl et al., 2015), are not considered. Other processes, such as the evolution of sulfate aerosol through microphysical processes, as nucleation, coagulation, or condensation~~, or evaporation~~, and sedimentation of particles with different sizes can play a role in our region of interest. However, comparisons of simulations and measurements show that these processes are not essential to study the ~~fate~~ development of sulfur emitted by Kasatochi in 2008 and Sarychev in 2009.

4.2 Sulfur transport

The Kasatochi eruption directly injected a large amount of SO_2 especially into the altitude region between 10 and 14 km (Table 1). Fig. 7 displays vertically resolved time series of SO_2 and liquid-phase H_2SO_4 mole-fractions~~,~~ for 30° N to 90° N, as measured by MIPAS (7a–c) and modelled by the CTM (7d–f), together with their sum (c and f). Both~~,~~ SO_2 and sulfate aerosol show a separation of the plume into a lower and upper part, in the measured and simulated data. While the lower part is removed relatively fast from the altitude range of observations, starting at 10 km, the upper part moves downward with time, following the Brewer–Dobson circulation. During the descent the sulfur concentrations are reduced and some parts of the sulfur reach 10–12 km after several months. ~~, and reaches 10 km after a few months.~~ As the descent is seen in sulfate aerosol and SO_2, it is not primarily driven by sedimentation. ~~In Fig. 6, the second increase of sulfur mass in the altitude region 10.5–14.5 km is also due to the descending part of the volcanic plume.~~ A similar pattern of subsidence was found by Andersson et al. (2015, Fig. 3 therein) when studying aerosol scattering measured by CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations). Additionally shown in Fig. 7 is the eruption of Sarychev in 2009. No separation of the plume is noticeable, vertically the sulfur is distributed rather homogeneously. In terms of downward transport from higher altitudes in the extra-tropics ~~from higher altitudes~~ and the removal from the stratosphere, SO_2 and sulfate aerosol from the Sarychev eruption evolve quite similar to the Kasatochi eruption.

Parts of the differences between the transport patterns after the eruptions arise from the injected SO_2 masses. In the case of Kasatochi the main part of SO_2 was injected to altitudes below 14 km (518 Gg or 77 % of the injected SO_2). It is transported downwards and out of the region studied here relatively fast and therefore only a minor part is reflected in the aerosol loading. In the case of the Sarychev eruption almost half of the SO_2 (367 Gg or 48 % of the injected SO_2) is injected into the altitude region above 14 km. It is available for conversion into sulfate aerosol for a longer period of time, as can be seen in the higher H_2SO_4 volume mixing ratios after the eruption of Sarychev, compared to Kasatochi. Model simulations with 'switched' SO_2 masses (mass of Kasatochi injected on the day and at the location of Sarychev, and vice versa), and a simulation with the SO_2 mass from the Kasatochi eruption injected at the location of Sarychev, reveal that the 'double-plume' that has been observed after the eruption of Kasatochi results from the combination of the vertical distribution of injected SO_2 masses and the prevailing transport after the 7 Aug 2008, the eruption date of Kasatochi, in the model driven by wind fields and heating rates. Neither of the simulations results in a comparable separation into an upper and lower part of the plume.

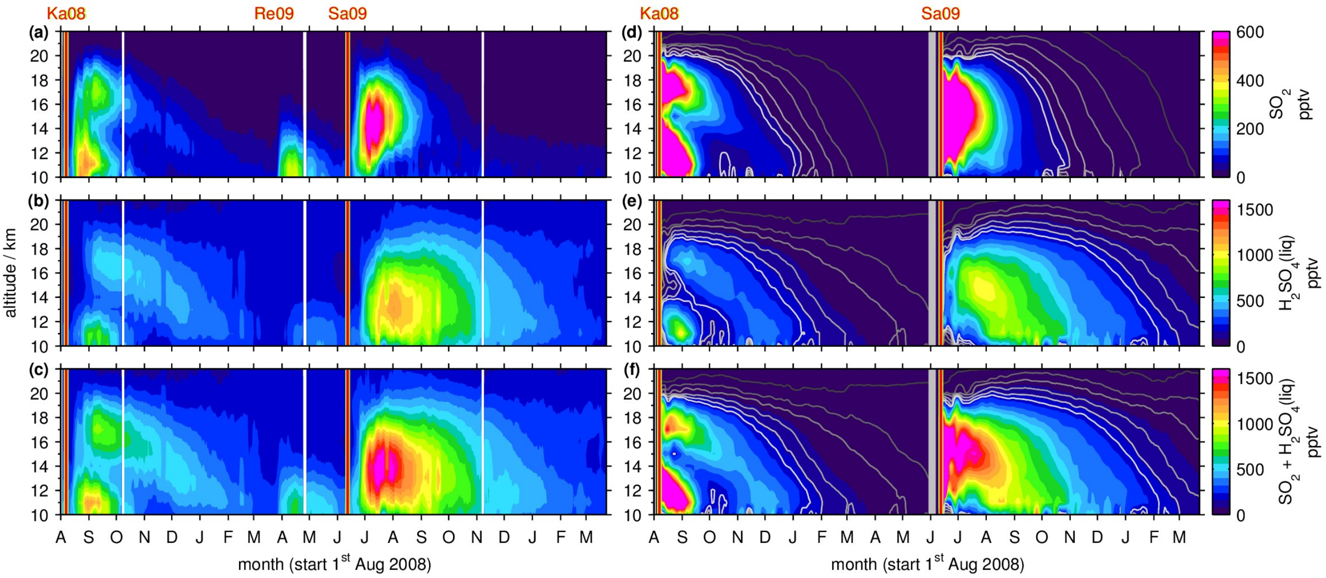

Figure 7. SO_2, liquid-phase H_2SO_4, and total sulfur (SO_2 + H_2SO_4) volume mixing ratios by MIPAS **(a–c)** and the CTM **(d–f)**. Vertically resolved time series of SO_2 and sulfate aerosol for 30° N–90° N, starting on the 1 Aug 2008 (area weighted 5d running means). Indicated are the days of the eruptions of the volcanoes Kasatochi in Aug 2008, Redoubt in Mar 2009, and Sarychev in Jun 2009, which were observed by MIPAS. Redoubt is not considered in the model. The colour scale is restricted to 0–800 pptv (SO_2), and 0–1,600 pptv (sulfate aerosol, and sum of SO_2 and sulfate aerosol). ~~Lower and higher~~ Exceeding values are assigned with the respective limiting value. For the CTM contour lines in dark to light grey represent 1, 5, 10, 20, 30, and 40 pptv in the case of SO_2, and 5, 25, 50, 100, 150, and 200 pptv for sulfate aerosol, and the sum of SO_2 and sulfate aerosol. For the CTM two simulations are shown, from August 2008 to May 2009, and from June 2009 to May 2010, separated by a bold grey line.

Figure 8 shows vertically resolved measurements and simulations of SO_2 and liquid-phase H_2SO_4 for 0° N–30° N. Some of the sulfur emitted at around 50° N reaches low latitudes. While the SO_2 is removed rather fast, compared to sulfate aerosol, the sulfate aerosol resides in the tropics for many months and moves upwards with time. The modelled sulfate aerosol with ~~a~~ an effective sedimentation~~-~~radius of 0.5 µm behaves in a similar way as the measurements, moving slightly upwards with time. In comparison, a simulation with ~~1 µm~~ an effective sedimentation radius of 1 µm shows a rather horizontal transport and faster removal, while the simulated lifting is stronger than in the measurements~~,~~ when not considering sedimentation (not shown here). Due to uncertainties in modelled ascent speeds (e.g. Pommrich et al., 2010; Liu et al., 2013), the particle radius that is most suitable to reproduce MIPAS measurements by CTM simulations is not necessarily the best estimate when performing similar analyses with different models or meteorological driving data.

In the MIPAS data of the tropics, where the tropopause height is relatively constant at around 16–17 km, a clear transition from elevated sulfur mole-fractions in the troposphere to lower sulfur loading ~~content~~ in the stratosphere is observed during times of weak volcanic influence (May–Jun 2009, Fig. 8c). The relatively high values at around 13–16 km in the measurements have already been noted in Fig. 5 and are supposed to only partly be connected to volcanic eruptions. A certain influence of elevated retrieved aerosol values due to cirrus clouds that have not been captured by the ice-filter (Sect. 3.1) is possible. To which extent the observed enhancements in the measurements (Fig. 8a–c) are caused by the eruptions of Kasatochi and Sarychev is not clear. In the case of Kasatochi model simulations suggest that enhancements are confined primarily to altitudes above approximately 16 km. Additionally to the tropical enhancements at 13–16 km, the eruption of Dalafilla in November 2008 overlays with the observed sulfur that has been emitted by Kasatochi. The CTM simulations of Sarychev indicate that sulfur observed at altitudes as low as 12 km can be attributed to the volcanic eruption.

Differences between the presented zonally averaged measurements and model results arise partly from the fact that MIPAS measurements are not uniformly distributed and data were filtered, and due to sparse data coverage in the case of the CTM up to an altitude of 12–13 km. Data are partly missing in relatively large areas, which may lead to biased zonal means. In the measurements, for SO_2 data are missing particularly in the tropics at altitudes below about 15/16 km and at higher altitudes (up to ~17 km) in the region of the Asian Summer Monsoon. In the case of measured sulfate aerosol data are filtered especially in the tropics at altitudes up to about 18/19 km and in the region of the Asian Summer Monsoon (up to ~20 km) and in polar regions entire profiles were filtered out due to PSCs. Especially after the eruption of Sarychev a higher sulfur content is simulated in the tropics compared to the measurements (Fig. 8) and enhancements are seen few days after the eruption. This results from a strong modelled meridional transport of SO_2 after this eruption. At about 12–16 km altitude the injected SO_2 reaches 15° N 7–8 d after the eruption. This strong southward transport early after the eruption is not reflected in the measurements, which are, however, partly missing in the tropics due to filtering.

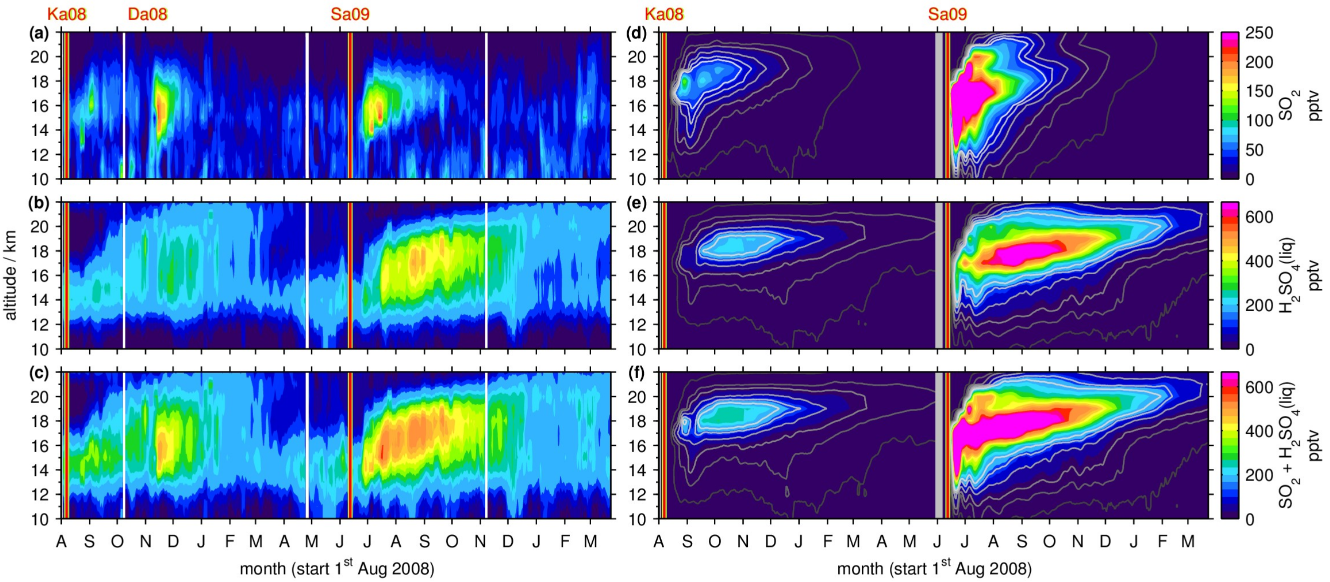

Figure 8. As Fig. 7, but for 0° N–30° N. The colour scales are restricted to 0–200 pptv (SO_2), and 0–800 pptv (sulfate aerosol, and sum of SO_2, and H_2SO_4(liq)). Additionally the volcano Dalafilla is indicated in Nov 2008, as it has been observed by MIPAS. It is not included in the model.

In Fig. 9 and 10 time series of latitudinally resolved mole-fractions show the transport of SO_2 and sulfate aerosol at different altitudes. We present time series of 5d running zonal mean mole-fractions for the Northern Hemisphere, at altitudes from 10 to 22 km, both for MIPAS measurements and CTM simulations. As the model has very poor coverage at 10 km, results are not shown for the model at this altitude. For the eruption of Kasatochi, the separation of the plume and downward transport of the upper part is notable at mid- to high-latitudes, most easily visible for sulfate aerosol (Fig. 10).

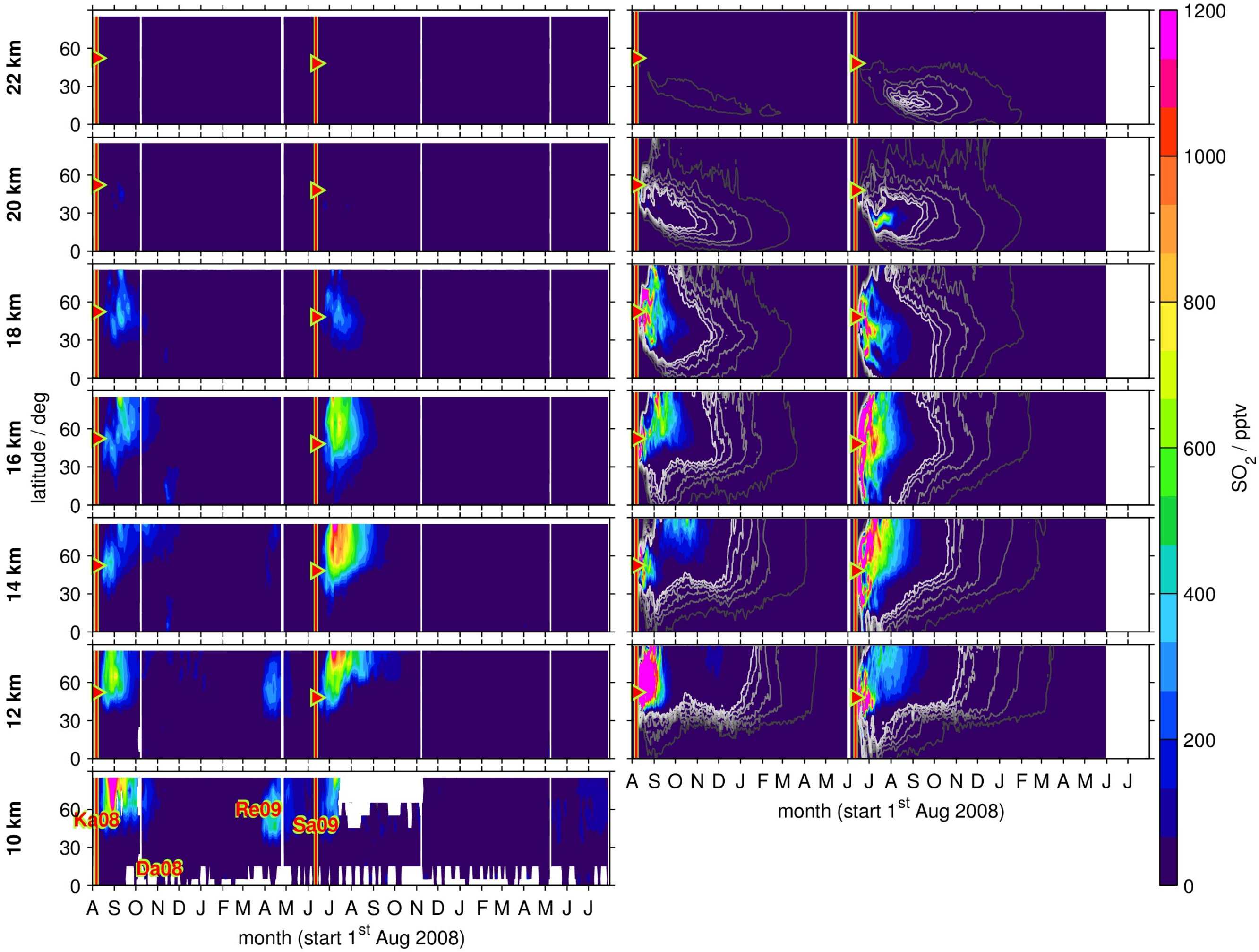

Figure 9. Time series of SO_2 from MIPAS measurements **(left panels)** and CTM simulations **(right panels)**. Shown are volume mixing ratios after the eruption of Kasatochi (7 Aug 2008) and Sarychev (12 Jun 2009), as zonal means for the Northern Hemisphere, at various altitudes (10 to 22 km). For MIPAS 5d running means for 10° latitude bins are calculated, while for the CTM daily values are shown for ~2.5° latitude bins. The colour code is restricted to 0–1,200 pptv. Values exceeding these limits are assigned with the limiting value, respectively. Time and location of the eruptions of Kasatochi and Sarychev are indicated by red triangles. (left) Indicated are the eruptions of Kasatochi and Dalaffilla in 2008, and Redoubt and Sarychev in 2009, that were observed by MIPAS. (right) The two simulations are separated by a grey line. Black to white contour-lines denote 1, 5, 10, 20, 30, and 40 pptv. Only Kasatochi and Sarychev are included in the model.

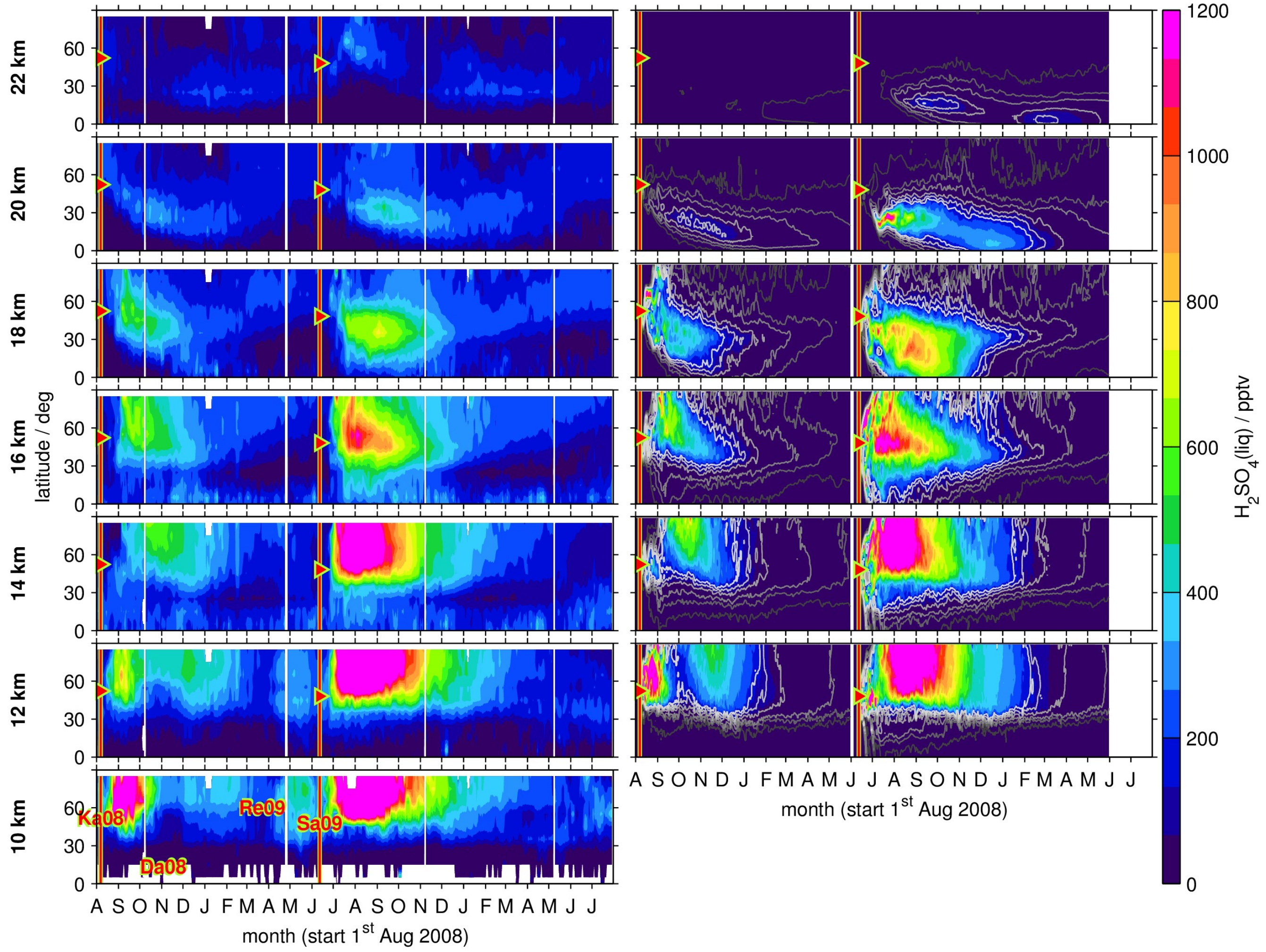

Figure 10. As Fig. 9, but for sulfate aerosol. (right) Black to white contour-lines denote 5, 25, 50, 100, 150, and 200 pptv.

Both~~,~~ in the measurements and simulations, most of the sulfur contained in SO_2 and sulfate aerosol stays north of 30° N at lower altitudes up to around 16 km (Fig. 9 and 10). Especially at low altitudes we find a mixing barrier at ~30° N, with a strong gradient between low values in the tropics and high values in the extra-tropics, which weakens towards higher altitudes. This gradient is due to the subtropical jet stream, and is most easily detectable in the contour lines shown for modelled liquid-phase H_2SO_4 (Fig. 10, right panel), but similar patterns are observed by MIPAS. At around 16–18 km, especially in the longer lived sulfate aerosol, this forms a “tongue” of relatively high mole-fractions, which persists over a longer period than in the surrounding latitudes. ~~An additional transport process starts at~~ At an altitude of about 18 km in the case of Kasatochi and ~16 km in the case of Sarychev~~ (Fig. 9 and 10)~~, and at altitudes above, a southward transport of sulfur is noticed (Fig. 9 and 10). At these altitudes, both the MIPAS measurements and CTM simulations show that sulfur from mid-latitude volcanic eruptions can reach the tropics, predominantly in the form of sulfate aerosol. In the tropics sulfur is then lifted in the 'tropical pipe' (denotation following e.g. Plumb 1996), and can reach the stratospheric 'overworld' (denotation following e.g. Hoskins, 1991), also seen in Fig. 8. Measurements of the stratospheric optical depth by the Optical Spectrograph and InfraRed Imager System (OSIRIS)~~/~~ onboard Odin also show that in the months after the eruptions of Kasatochi in 2008 and Sarychev in 2009 their impact extended to lower latitudes (Bourassa et al., 2012). Wu et al. (2017) studied the equatorward dispersion of the Sarychev volcanic plume together with the influence of the Asian summer monsoon on the transport pattern. They find~~,~~ that at 360–400 K potential temperature~~,~~ the southward transport was primarily caused by anticyclonic Rossby wave breaking, intensified by the Asian summer monsoon during Northern Hemisphere summer. Above 400 K less aerosol is transported into the tropics. They further show an 'aerosol hole' in the anticyclone, surrounded by aerosol-rich air. Following Wu et al. (2017)~~,~~ a strong subtropical jet in combination with weak Rossby wave breaking events would hinder the southward transport of the volcanic plume during winter conditions. ~~The weaker southward transport in the case of the Kasatochi eruption that starts at higher altitudes, compared to the Sarychev eruption, could be due to the eruption~~ Compared to Sarychev, the southward transport of the Kasatochi eruption plume is weaker and initiates at higher altitudes. This might be explained by 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. In model studies of the Sarychev eruption, Haywood et al. (2010) find that sulfate aerosol is transported around the entire globe in around 14 days. We see that the bulk of the aerosol that moves southwards reaches the equator about 2 to 3 months after the volcanic eruption. To some small extent this sulfur crosses the equator and can thereby ~~hereby~~ influence the sulfur loading ~~content~~ of the Southern Hemisphere (see also Wu et al., 2017). Generally, similarities in the geographic pattern between Kasatochi in 2008 and Sarychev in 2009 have also been noticed by Haywood et al. (2010), caused by the agreement between the season~~ of the eruptions~~, the injection altitude and latitude of the eruptions. The model results and MIPAS measurements of SO_2 and sulfate aerosol presented in our study confirm similarities in the transport patterns of sulfur after the volcanic eruptions of Kasatochi in August 2008 and Sarychev in June 2009, and a southward transport of the volcanic plumes, towards the equator, where sulfur can then ascent in the Brewer–Dobson circulation.

5 Discussion and conclusions

In this study a new ~~dataset~~ data set of MIPAS/Envisat global aerosol volume densities, also converted into ~~and~~ liquid-phase H_2SO_4 ~~VMR~~ volume mixing ratios ~~distributions~~, is presented for 2005 to 2012, covering the altitude range of 10 to 30 km, with up to 1,300 profiles per day, derived from single limb-scans. The MIPAS aerosol volume densities have been corrected for ~~possible instrumental radiance baseline offsets by comparison to coincident balloon-borne in situ observations from Laramie, Wyoming~~ a positive bias in comparison to coincident balloon-borne in situ observations from Laramie, Wyoming. This bias is supposed to be caused by instrumental radiance baseline offsets. With absolute differences below ±0.003 µm^3 cm^{-3} at 20–25 km the bias corrected MIPAS profiles compare well with the in situ data. The strongest variability in the MIPAS sulfate aerosol is caused by various volcanic eruptions. Liquid-phase H_2SO_4 patterns from MIPAS are in general agreement with MIPAS SO_2 profiles from single limb-scans during volcanically perturbed and quiescent periods.

In a case study~~,~~ we investigate the ~~fate~~ evolution of volcanic sulfur after two major mid-latitude volcanic eruptions of the last decade (Kasatochi in 2008 at 51.2° N, and Sarychev in 2009 at 48.1° N) by combining this new ~~dataset~~ data set with simultaneously observed profiles of SO_2 from the same instrument with the help of CTM simulations. Liquid-phase H_2SO_4 derived from the MIPAS aerosol retrieval is not only qualitatively, but also quantitatively consistent with the MIPAS SO_2 observed after the two volcanic eruptions. One of the advantages of deriving aqueous H_2SO_4 and SO_2 from one instrument is that no sampling biases occur due to different geolocations and measurement times. Remaining ~~sampling biases~~ discordances are caused by different filter methods, which depend partly on the retrieved species. The ~~datasets~~ data sets provide a valuable tool for further analyses of the stratospheric sulfur loading ~~content~~. The new H_2SO_4 aerosol observations enable us to further constrain the total sulfur emitted into the stratosphere by the Kasatochi and Sarychev eruptions and to revise our previous estimates that were based on SO_2 observations only. The new estimates are 677 Gg SO_2 in the case of Kasatochi and 768 Gg SO_2 in the case of Sarychev that were injected into the altitude range 10–19 km. The decay of SO_2 after the volcanic eruptions and the formation of sulfate aerosol are consistent with known SO_2 chemical lifetimes due to reaction with OH, under OH background conditions (modelled OH climatology without any feedbacks between sulfur species and OH concentrations). While sedimentation of sulfate aerosol does play a role, the ~~residence time~~ decay of sulfur in the mid-latitude lower stratosphere following the volcanic eruptions of Kasatochi and Sarychev is dominated by advective transport and transport by the Brewer–Dobson circulation. Sensitivity simulations with the CTM with different effective ~~aerosol~~ sedimentation radii indicate that the observed ~~decay of~~ sulfate aerosol is best described by sedimentation of aerosol particles with an effective radius of about 0.5 µm. Most of the sulfur emitted by the two volcanic eruptions resides in the extra tropical lowermost stratosphere where ~~were~~ it is transported downward across the tropopause. However, at higher altitudes (at about 16 to 22 km) parts of the volcanically emitted sulfur from these Northern Hemisphere mid-latitude volcanoes is transported equatorwards where it is lifted in the 'tropical pipe' into the stratospheric 'overworld' and even enters the Southern Hemisphere.

Our findings of the residence time and transport pathways of enhanced sulfate aerosol in the mid-latitude lower stratosphere have implications for the forcing of surface climate by moderate sized mid-latitude volcanoes and proposed geoengineering schemes. Sulfur injections into the lowermost stratosphere at mid-latitudes can affect not only the mid- to high-latitudes of the respective hemisphere, but are potentially transported towards the tropics, where they can undergo uplift and further transport by the Brewer–Dobson circulation, and can thereby ~~hereby~~ reach the other hemisphere.

6 Data availability

The MIPAS ~~datasets~~ data sets for aerosol volume densities and liquid-phase H_2SO_4 mole-fractions are available upon request from the authors or at http://www.imk-asf.kit.edu/english/308.php. Model results are available upon request from the authors.

7 Appendix

Method of increasing areas for zonal averages:

To reduce biasing of zonal averages due to non-uniformly distributed data, we use a method of increasing areas. It is based on the horizontal grid of our Chemical Transport Model, which has a resolution of ~2.5° latitude x 3.75° longitude. For MIPAS daily arithmetic means are calculated for these ~2.5° x 3.75° boxes. These are then averaged to 32 boxes of ~ 10° x 11.25°. The area is further increased longitudinally by a factor of 2 in each step, while the number of boxes decreases by the same factor (32 x 11.25° → 16 x 22.5° → 8 x 45° → 4 x 90° → 2 x 180° → 1 x 360°). The result is not changed, compared to normal averaging (sum of values divided by number of values), when an equal number of values is available per ~ 2.5°x3.75° grid-cell, which is the case for the CTM, in altitudes above 13 km. By interpolating the model results to a vertical grid with 1 km resolution, starting at an altitude of 10 km, we find missing values up to 13 km. Therefore the method of increasing areas for zonal averages is applied to the simulated data in these altitudes as well. As the surface decreases with increasing latitude, the latitude bin is chosen to be 11.25° and not broader, as increasing latitude bins can give a too high weight to values corresponding to relatively smaller areas.

8 Author contributions

A. Günther developed and performed the model simulations, wrote most of the paper and conducted most of the analyses. M. Höpfner developed the MIPAS aerosol retrieval and provided the retrieval sensitivity studies and error estimations, and their description (Sect. 3.1). B.-M. Sinnhuber provided advice with the development and analysis of chemical transport modelling. T. von Clarmann, G. Stiller and M. Höpfner provided advice for the analyses of MIPAS data. S. Griessbach provided the flags for the MIPAS ice and ash filter. T. Deshler provided the balloon-borne in situ data and their description (Sect. 2.2). All authors contributed to the discussion of the results.

9 Competing interests

The authors declare that they have no conflict of interest.

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