Interactive comments on

Model simulations of the chemical and aerosol microphysical evolution of the Sarychev Peak 2009 eruption cloud compared to in-situ and satellite observations

by Thibaut Lurton et al.

Authors' foreword:

We wish to thank both referees for their comments on the manuscript.

Please find below our answers to their remarks: their original comments are typed in italics, and we address our answers following each point raised.

Answers to Anonymous Referee #1

This is a great study. The importance is clear. The authors have done an excellent job with their analysis - I'm quite impressed with the care and thoroughness they have applied to this research. I only have a few minor comments.

Page 2, lines 24-25

I think you're conflating two issues here. I agree that small eruptions at high latitudes would likely have impacts that are confined to one hemisphere. I also agree that large eruptions in the tropics would likely impact both hemispheres. This is not an either-or. What about small eruptions in the tropics or large eruptions at high latitudes? This sentence needs to be written more carefully.

We agree with the reviewer, as we forgot to mention small tropical eruptions (e.g. Soufrière in 2006 or Kelud in 2014) which affected at least one of the hemispheres, depending on the QBO phase, and likewise, we do not mention major eruptions having affected one hemisphere only. We therefore rewrote the concerned lines:

"However, they typically have a much-reduced effect on climate and atmospheric chemistry compared to large-magnitude eruptions (Oman et al., 2005; Kravitz et al., 2010). In general a smaller mass of SO_2 is injected and oxidized to sulfate aerosol. Also, by injecting to lower altitudes, the emissions from moderate-magnitude eruptions are more susceptible to removal by stratospheric-tropospheric exchange processes. Nevertheless..."

Page 5, line 17

Did this overly dilute plume affect your results?

The question is about the initial dilution of the injected SO_2 into the model grid, which is an unavoidable consequence of global model simulations using large grids (few degrees) as for other studies, e.g. Haywood et al. (2010). Nevertheless, we find good agreement in our model-observation comparisons of aerosol several months after the eruption. This indicates that the initial dilution does not seem to have important impact on our comparisons over those time-spatial scales. To fully test the reviewer's question would require modelling over a finer grid. That is beyond the scope of our study, but is indeed of interest for future studies as we can anticipate availability of more powerful computational resources as well as new upcoming satellite data at higher resolution.

Page 5, general

You don't talk too much about the effects of the vertical distribution of the aerosols.

We have added a sentence to line 18:

"The vertical distribution of our SO_2 injection follows previous model studies (e.g. [Haywood, 2010]). It is a somewhat coarse approximation given that O'Neill et al. (2012) report lidar observations of fine-scale aerosol layers shortly after the eruption. Nevertheless, these were subsequently observed to collapse into a single layer in the lower stratosphere. For the magnitude of the SO_2 injection we use a revised estimate that contrasts to previous studies, as discussed below."

Page 8, line 7

Maybe I'm misinterpreting your colocation metric, but it doesn't look like the plume is "reasonably well simulated" by the model. Some clarity is needed here.

The colocation metric we used is as a matter of fact the Pearson's correlation coefficient, which takes into account both differences in amplitude and location. Another alternative would have been to use the Spearman's rank correlation coefficient, which operates on ranked variables, and therefore is less sensitive to strong outliers and does not depend on a linear relationship between the variables. Though this latter coefficient is more flattering in terms of matching scores, we chose to retain our first approach.

We consider our plume reasonably well simulated by the model, as compared visually (Figure 1) to figures from previous studies such as Haywood et al. (2010) (they do not report colocation metrics to compare quantitatively). This is further confirmed by our analysis of Northern Hemisphere SO2 burden in Figure 2.

Figure 2 and surrounding analysis

Mills et al. (2016) show that WACCM+MAM3 simulates Pinatubo really well. This is using the same model (albeit with CARMA instead of MAM), but there are some discrepancies in Figure 2. Can you say more about why?

The question raised by the reviewer is tricky to address. It is difficult to compare different model configurations and associated simulations conducted for two different eruption events which correspond to different dynamical contexts (i.e. latitude and altitude range of injection) and aerosol microphysical properties (i.e. size distributions). Perhaps the size distribution shape corresponding to the Pinatubo aerosol are sufficiently addressed with a modal aerosol scheme. However, our Fig. 2, displaying SO_2 temporal evolution is not directly comparable to Fig. 5 from Mills et al., which presents AOD. Also, one should note that conversely to our Fig. 2, Fig. 5 presented by Mills et al. is in log-scale so it is somewhat complicated to definitely infer a very good agreement between MAM3 and observations in this study.

Answers to Anonymous Referee #3

In this study the authors use CESM1(WACCM)-CARMA simulations to show the impact of volcanic HCl on volcanic SO2 life time and on ozone and NO_x depletion. Further the authors compare their simulations with IASI SO_2, balloon-borne particle measurements, and OSIRIS SAOD. Special emphasis was put on the comparison with OSIRIS data accounting for the instrument's limitations.

Fundamentally the study is sound. I recommend it for publication in ACP following revisions suggested below.

Major comments:

page 2 line 33:

Although the IASI SO2 retrievals are sound and precise, I'm not convinced that they should be the first choice the estimate injection heights. ACE (Doeringer et al, JGR, 2012), CALIOP (e.g. Solomon et al., Science, 2011), MIPAS (Höpfner et al., ACP, 2013, 2015) and the ground based lidar measurements you mentioned clearly show that a significant part of the Sarychev SO2 was injected above 15 km.

page 5 line 14-16:

Although you justify your choice of a Sarychev injection on 15 June only into altitudes between 11 to 15 km in the next paragraph there are also studies demonstrating that a substantial amount of SO2 reached higher altitudes (ACE (Doeringer et al, JGR, 2012), CALIOP (e.g. Solomon et al., Science, 2011), MIPAS (Höepfner et al., ACP, 2013, 2015)). Images of different instruments (e.g.

http://sacs.aeronomie.be/nrt) show that there was a significant amount of SO2 injected before the 15th and a very recent study in this journal provides an emission time series that placed the onset of the strongest eruption phase in the afternoon of 14 June (Wu et al., ACPD, 2017). Also Levin et al. (2010) found the onset of the second strongest eruptions on 14 June at 18:50. I suggest taking this into account. Please see also further minor comments on this aspect.

IASI altitude retrievals have been shown to be accurate in general within 1—2 km (e.g. Clarisse et al. (2014), Carboni et al. (2016)), so that we believe that IASI can be used to make statements on the injected altitude. For the early Sarychev plume, some SO_2 is measured up to 20 km, but the majority (> 95%) of the SO_2 mass was found to be below 15 km. This last statement is true for both the retrievals presented in Carn et al., 2016 and Carboni et al., 2016, which each use an independent altitude retrieval algorithm.

Other instruments (limb/occultation/lidar) have indeed a more inherent sensitivity to altitude, although limb/occultation measurements also have limitations in their vertical resolution. However, we disagree that these measurements unambiguously show that a “significant part” of the plume is located above 15 km in the early plume. Part of the problem is that some of these instruments have a very limited coverage, and that therefore measurements are often reported within the aged air masses, which can undergo significant vertical transport over time (Vernier et al., 2011), and thus no longer provide information on the injection altitude.

Here we summarize the observational evidence from the papers that the reviewer mentions:

MIPAS: Höpfner et al. give the following numbers 888 kT (10—14 km) / 542 kT (14—18 km) / 44 kT (18—22 km). It is unclear over which time period these measurements were gathered.

CALIPSO measurements of aerosols: Solomon et al, 2011, Fig. 1, show the SR@532nm between 17 and 21 km, and indeed shows enhancement in the wake of the Saychev eruption. It is however not possible to conclude from this plot that a significant part of SO_2 was injected at those altitudes.

Prata et al., ACP, 2017 (part 4.2) indicate that no CALIOP data is available for the 12—14 June period.

Doeringer et al., 2012, Fig. 6, shows a peak in the median atmospheric extinction in July 2009 at 13 km, which quickly drops off below 9 km and above 15km. Figure 7 shows similar result, with a SO2 tail at 16,17 km altitude on 14 July. Fig. 8 shows a similar profile for SO_2.

A trajectory-model approach by Wu et al. (2017) finds the largest SO_2 injection occurred between 12 and 17 km [Günther et al., 2017].

We must also emphasise on the fact that our model vertical resolution as far as injection is concerned is 1 km. Therefore, the precision in the injection altitudes is somewhat coarse, and subject to some trade-off. A detailed study of the complexities of the injection, tracing the horizontal and vertical transport of fine-scale plume filaments is not the goal of our study at the resolution of our global model (~2 degree grid, ~1 km vertical resolution) that focuses on a detailed chemistry and aerosol microphysics. Such efforts are rather suited to trajectory-dispersion models e.g. the recent study by Wu et al., ACP (2017) of transport pathway of the Sarychev SO_2 emission and sulfate aerosol from the extratropical lower stratosphere to the tropical tropopause layer (TTL).

A visual analysis of Figure 2 of Wu et al. (2017) depicts some injection > 15 km in afternoon-evening of 14 June. To estimate the load requires looking at the concentration multiplied by the area (in altitude-time axes) of the plot. This suggests that between 12 and 15 km altitude is a suitable approximation for the majority of the emission, as needed to investigate large-scale evolution patterns. The studies of Wu et al. and Gűnther et al. (2017) indicate that emissions at higher altitude could particularly affect transport of plume to southern latitudes. Therefore our choice of injection altitude may limit our modelling of this aspect, but it is not the focus of our study that focuses on the NH evolution and observations at mid and high latitudes.

On the comment of the eruption time: IASI measures about 100 kT on the evening orbit of the 14^{th}.

So indeed there was a non-negligible amount of SO_2 emitted before the 15^{th}, but according to IASI most of it was clearly erupted on the 15^{th} / early 16^{th}.

Last, we point out the fact that our study had to keep a relative consistency with the previous studies to which it is compared throughout. This advocates for the injection altitudes and times we chose, which are comparable to those of the papers we use as points of comparison.

Minor comments:

page 2 line 15:

What do you mean by "global visible AOD was enhanced by up to 0.15"? Is 0.15 a factor or the AOD?

It is the AOD. We replaced the text by "global AOD (in the visible) was enhanced, reaching up to 0.15" in the manuscript.

page 2 line 19:

Please consider also the Arctic, e.g. Tilmes et al., ACP (2008), as the Sarychev eruption that is discussed here affects the Arctic.

We replaced the clause "Antarctic ozone hole" by "polar ozone holes", and made reference to Tilmes et al., 2008.

page 2 line 24:

Can you add references?

This part of the text was rewritten following Referee #1's remark.

page 2 line 31: Only SO2 and HCl or also ash?

Indeed, some ash was injected, though it was not prescribed in our model runs. Mention of ash has been however added to the concerned sentence. We discuss later that we do not consider ash in this study. Here we are focused on SO_2 (and co-injected HCl mentioned as it is an aspect of our study).

page 3 line 8: Here I'd like to add that a very recent study in this journal found simulations with a "sedimentation radius" of 0.5–1 µm to match best with observations (Günther et al., ACPD, 2017).

We added the reference within the manuscript.

page 3 line 10-15:

The reff derived from ACE remote sensing measurements was also 0.1 – 0.3 µm.

We have added the sentence:

“Further evidence for a larger particle size comes from effective radius estimate of 0.1-0.3 µm derived from satellite-based observations on month after the eruption (Doeringer et al., 2012) and a particle “sedimentation radius” of 0.5 – 1 µm from a model sensitivity study (Günther et al., ACPD, 2017).”

page 5 line 28, 30, 32:

What are the uncertainties of the SO_2 burdens? Do they agree within their uncertainties?

A typical uncertainty on SO2 burden retrievals using the [Clarisse et al., 2012] algorithm would be 10—20 %. Considering the highest uncertainty, the 0.9 Tg estimation still stands out as being different from 1.2 Tg. This was added to the manuscript (see remark concerning p.8, l.5—11).

page 6 line 1:

What is the uncertainty of the HCl injection?

Carn et al., 2016 mention HCl at 7—9 ppbv, compared to 529 ppbv SO_2, but do not provide any other figure to derive uncertainty, that is difficult to quantify as discussed in their text. As far as our study goes, we tested the sensitivity to the presence of HCl, and therefore our results should be considered as a very first investigation of the impacts of the co-injection of HCl with SO_2 from the Sarychev eruption, using the best-available estimate from Carn et al. (2016).

page 6 line 18/19:

How did you determine the tropopause? What is the uncertainty of the tropopause altitude?

The model tropopause was used (see further details below). Therefore we cannot properly state a tropopause altitude uncertainty; the tropopause height is self-consistent with the model.

page 6 line 21-29:

How do you justify a comparison with SO_2 column data, while neglecting all injections below 10 km in the simulations?

Please refer to the answers to the major comments concerning injection heights. We have no precise number concerning injection below 10 km. Furthermore, as the plume eventually goes down in altitude, it is useful to monitor the column data. We here focus on volcanic sulfuric acid particles in the stratosphere. The tropospheric lifetimes of sulfate aerosols and SO_2 are shorter, due to wash-out processes and clouds.

page 7 line 6:

Please provide a valid URL for the STAC data in indicate your last access (for all urls). After a short search I found the following site claiming to provide STAC data, but ended up at blank pages or 404: http://cds-espri.ipsl.upmc.fr/etherTypo/index.php?id=667L=1

The problem was acknowledged, and the ESPRI website team was notified accordingly.

page 8 line 5/11:

I suggest considering adding the IASI SO2 retrieval threshold information and its altitude sensitivity range to the description of the data set in Section 2.2.

We added the following sentence to Section 2.2:

"IASI retrievals have a typical altitude sensitivity of 1—2 % km. For the precise Sarychev eruption retrievals, SO2 loads can be expected to have a 10—20 % uncertainty."

page 8 line 8: How do you know that this is due to SO2 injected before the 15 June?

We toned down our assertion, rewriting the sentence as follows: "this is likely to be due to our simulation not accounting for the small amount of SO2 that was emitted before the main eruption".

page 8 line 11-16:

Which model output time did you use for the comparison? The same as the measurement time of each orbit?

No average was performed, in order not to over-dilute the signal. We considered two instantaneous outputs per day, at 0:00 and 12:00.

page 8 line 24 - page 10 line 6:

This part was confusing. I'd suggest reordering and rewording. E.g. present your simulation results first, second your simulation results but with IASI detection threshold, third Haywood model and IASI data. Also consider moving the information on the IASI SO2 retrieval threshold to Section 2.2.

The text is rearranged as follows:

"Fig. 2 shows the modelled northern hemispheric SO_2 burden in Tg, calculated by integrating the model anomalies from CESM1(WACCM) simulations with SO_2 injection only and with SO_2 and HCl co-injection (anomaly denotes a "volcano-on" simulation from which the "volcano-off" control run has been subtracted). Two adjusted CESM1(WACCM) model results are also presented that only include data over columns with > 0.3 DU SO_2 to enable a better comparison to the IASI observations. Alongside is shown the observed evolution in northern hemispheric SO_2 burden derived from the IASI retrieval by Clarisse et al. (2012) (that has a lower threshold of around 0.3 DU, see Methods 2.2). Finally, we also show the northern hemispheric SO_2 burden as simulated using the HadGEM2 model (Haywood et al., 2010), and the IASI retrieval reported in that same study, both of which estimated 1.2 Tg SO_2 injection in contrast to the revised IASI analysis (Clarisse et al., 2012) that yielded 0.9 Tg SO_2 used in our study."

We add the following sentence to page 6, line 28:

"For this comparison we use the IASI retrieval of SO_2 by Clarisse etal. (2012). The IASI dataset and retrieval algorithm used for this precise eruption can be considered as showing a lower threshold of around 0.3 DU."

page 9 figure 1:

Would your comparison improve if you use 18:00, which is right in the middle of the post-meridiem period, instead of 00:00 model output? What do you mean with "this precise IASI retrieval"? particular? I suggest reducing the number of colors in this figure. I cannot distinguish the many shades of red, blue, and green in the figures. I assume that 7 distinct colors are enough. This type of figure I've just seen in Wu et al. 2017 for a comparison between AIRS data and model output. I suggest a comparison.

The IASI data are averaged over the AM and PM local periods. We use instantaneous snapshots of the model as a comparison, at 0:00 and 12:00, which is (as the referee points out) not a perfect match, but in our opinion remains sufficient to demonstrate the good behaviour of the model in comparison to the observations.

The clause "this precise IASI retrieval" refers to the possible different computation methods in the IASI retrievals, which can vary. As for the number of colours, we choose not to change it in order to keep a useful high dynamic range on the onset of the eruption.

The comparison with Wu, 2017 indeed shows good agreement. We add, to conclude paragraph 3.1: "The spatial and temporal evolution of the plume in our study is consistent with the results of Wu, et al. (2017), where AIRS data are presented along with simulations by a particle dispersion model."

page 11 line 1.

What do you want to say? Do you mean all model runs or only the "unadjusted" model runs?

We mean all the unadjusted model runs. This was clarified in the text.

page 11 line 2-5.

This sentence is confusing. Please clarify.

We propose the following modification to the text:

"A second notable result is that all the unadjusted model outputs overestimate the SO_2 burden following the eruption compared to IASI measurements. The HadGEM2 model SO_2 exceeds the Haywood et al. (2010) IASI observations for the whole period. The unadjusted CESM1(WACCM) outputs also exceed the Clarisse et al. (2012) IASI observations after a few days. This behaviour contrasts with the two adjusted CESM1(WACCM) model outputs that correct for the 0.3 DU SO_2 lower value of the particular IASI retrievals used. The adjusted CESM1(WACCM) model outputs remain in close agreement to the observed post-eruption SO2 burden for the first 1–2 weeks, after which the model-simulated SO_2 burdens decline more rapidly than the IASI 2012 observations. This evolution can be expected: a greater dispersion in the 2°×2° model grid cells than in reality (and than observed by the IASI footprint of tens of kilometres), would cause an underestimation of the model SO2 burden compared to IASI. This effect will become more pertinent with dilution over time as the SO2 column approaches the 0.3 DU limit."

page 11 line 13/14:

Do you mean the maximum on 0.9 Tg here? Please clarify.

The text was changed to:

"For these calculations we choose the SO_2 burden maximum as the initial value (0.9 Tg in our study)."

page 11 line 15-page 12 line 2:

Jumping between your results and the findings of Haywood confused me. Consider presenting your results first and compare then with the results of Haywood.

We propose the following reworked text:

"The e-folding time-constant for SO_2 is approximately 17.0 days for the simulation including HCl, about two days longer than the approximately 15.0 days for the simulation that was run without HCl. When these CESM1(WACCM) model outputs are adjusted to correct for the 0.3 DU SO_2 lower value of the particular IASI retrievals used they yield e-folding time-constants of 11.5 and 10.0 days, respectively. For the IASI SO_2 retrieval of Clarisse et al. (2012) we calculate 12 days, i.e. very similar to the adjusted model simulation with SO2 and HCl co-injection (11.5 days). For comparison, Haywood et al. (2010) report that the HadGEM2 model yields a 13–14-day SO_2 e-folding time (assuming a higher SO_2 injection of 1.2 Tg and no HCl co-injection). Regarding IASI observations, Haywood et al. (2010) report an IASI SO_2 e-folding time of 10–11 days, whilst using our method we calculate 9 days for the IASI retrieval of 2010. This is summarised in Table 2."

page 12 table 2:

What is the significance of your e-folding time? All results are presented as integers, but the one for your model run with HCl and IASI detection threshold says 11.5 days.

We added a decimal naught in our results in order to clarify the precision of the computed e-folding times.

page 12 line 16:

Can you quantify the "good general agreement"? Is the agreement in the upper panel of Figure 4 within the error of the OPC and the uncertainty of your volcano-off simulation?

page 13 figure 4:

I suggest to add the measurement errors (that are given in Section 2.2) to the OPC data. Without them it is really difficult to judge if the simulations and observations agree quantitatively within their errors on the logarithmic scale. Further, can you indicate the uncertainty range of the simulations?

We added the OPC error-bars on Fig. 4. The "uncertainty" of the model runs is something very difficult to estimate: it would depend on the dynamical processes, chemical processes, resolution used, etc. and would be very difficult to estimate on single-configuration runs. We did not carry out ensemble simulations, which were out of the scope of this study. The resolution we used was somewhat classical: it would have been cumbersome to lower it, and furthermore it is easily comparable to those of the studies we used as references.

page 14 line 2:

In Section 3.2 (page 10) you mentioned that your model is too disperse. It seems to me that here it is the most likely source of error. I'd suggest to compare with your IASI observations as in Fig. 1 and add this to Figure A1. You could also compare with Wu et al., ACP, 2017 and discuss.

IASI only sees SO_2, not sulphate particulates, and it is too late in time to compare SO_2 over Laramie on the dates considered (SO_2 is too dispersed to be observed). As for the comparison with Wu et al., 2017, 22nd June is not readily readable on Wu et al.'s plot, and furthermore it is an SO_2 map, not sulphates. Instead we do now earlier mention the SO_2 results of Wu et al. (2017) in relation to our Figure 1.

page 14 line 13:

How do you know that the CN particle mode has ever been different from the volcano-off simulation over Laramie?

We here refer to the simulations, and the CN mode has been different throughout time. We reformulated the sentence as:

"... indicating the progressive return of the simulated concentrations to background conditions for this size range".

page 14 line 25-30:

You discuss the discrepancies between the measurements and your model results at altitudes below *and above your injection height. I assume that not injecting SO2 below 10 km and above 15 km also contributes to the differences. I suggest to add this to the discussion.*

Concerning the possible injection of SO_2 below 10 km of altitude, we disagree with the referee: because of wash-out processes, it is impossible to maintain a tropospheric signal after such a long period of time. As for the injection above 15 km, please refer to our answers to the major comments; a substantial injection at these altitudes remains to be clearly proven (see e.g. Mattis et al. (2010), Doeringer et al. (2012), Jégou et al., (2013)). We add text to to the manuscript discussion-conclusion about uncertainties in injection altitude, see our response below to the comment for *page 25 line 9-11*.

page 15 figure 5:

Why are you using different colors for similar size bins (e.g. top: 885 nm is orange, bottom, 850 nm is red)? Why are there 3 size bins below 440 nm on 18 May 2010 but the other profiles start with 440 nm? I suggest to merge the lowest size bins for 18 May to make it comparable to the measurements in August. Please indicate the measurement and simulation uncertainty. On a logarithmic scale it's really hard to tell if there is a good agreement. Please also optimize the colors. In the 2 bottom panels there are two indistinguishable green lines.

Size bins for STAC change according to the calibration process, and therefore they can differ for different flights. Our principal point is the consistency between measurements and model computations. We added the error values on the total STAC counts (+/-6%) in the legend; for clarity reasons, we chose not to add them on the plots directly, which would make the figures unreadable.

page 16 line 1:

Has there ever been a comparison between STAC and OPC that might explain the difference?

Yes, indeed, Renard et al., Applied Optics, 2002, provides such a comparison. This paper shows a good accordance between the instruments.

Jean-Baptiste Renard, Gwenaël Berthet, Claude Robert, Michel Chartier, Michel Pirre, Colette Brogniez, Maurice Herman, Christian Verwaerde, Jean-Yves Balois, Joëlle Ovarlez, Henri Ovarlez, Jacques Crespin, and Terry Deshler, *Optical and physical properties of stratospheric aerosols from balloon measurements in the visible and near-infrared domains. II. Comparison of extinction, reflectance, polarization, and counting measurements*, Appl. Opt. 41, 7540-7549 (2002).

We added a sentence p. 7, l. 18: "Note that both STAC and University of Wyoming OPCs have been compared in Renard et al. (2002)."

page 16 line 1-3:

Does your model simulation suggest coagulation, condensation and sedimentation? What about transport to lower latitudes and dilution between August and November? What is the sedimentation speed and distance of e.g. 0.5 µm particles over 3 months? Shouldn't they show up at a lower *altitude in the OPC data then? Please substantiate your explanation.*

The model does indeed accounts for coagulation, condensation and sedimentation. It is very difficult to address the referee's question, since all processes (transport, coagulation, sedimentation) act together within the model. It appears hard to disentangle each of them, unless one carries out separate simulations activating processes one by one.

To answer the example question: the sedimendation speed and distance vary with the altitude of injection. Hamill et al. (1997) state that a 0.5 µm particle injected e.g. at 22 km of altitude has a sedimentation speed of 0,005 cm/s, i.e. 133 m/mth, which is slow.

We added reference to the transport and dilution processes in the revised version of the manuscript:

"This indicates the result of coagulation, condensation, sedimentation, and transport and dilution processes"

page 16 line 32/33:

How do you estimate the local tropopause? Do you use the thermal or dynamical tropopause? Which PVU threshold? What do you use at the pole/equator? What is the uncertainty of your tropopause? Please provide details.

According to WACCM's documentation, the local tropopause is calculated using the WMO lapse rate definition (thermal definition).

We erroneously stated that dynamical tropopause was considered: this was corrected in the revised version of the manuscript.

Tropopause levels are aligned to the altitude levels of the model, so that the possible error follows steps of ~1 km in altitude.

page 17 figure 6:

As I understood, the main purpose of this figure is to compare the STAC measurements with the model simulations, I suggest to select a smaller range on the y-axis so that it fits to the STAC data (e.g. dN 1e-3-1e2, dV, 1e-15-1e-12, dV 1e-21-1e-17). In the present figure I can only see that the no-volcano runs do not fit. Further, I consider error bars on the STAC measurements helpful.

We reworked the figure as suggested, with the y-axis ranges given. The error bars were not included on the figures for the sake of clarity, but mention to an error on the totals of +/-6% was added to the legend.

page 18 line 4-6: Why don't you rely your Zmin(Φ,λ,t) not solely on your analyses of 2009 shown in Fig A2? I consider a 2009 histogram more appropriate than a 2012 histogram with corrections.

Actually, the histogram considered was indeed over the period 2009—2010. The manuscript was corrected accordingly.

page 18 line 6.

Ok, it's dynamical tropopause. Which PVU is your tropopause? What do you use in the tropics/at the equator? 380 K? Thermal tropopause? What is the accuracy of your tropopause? Please provide details.

Please refer to our answer to a previous point concerning page 16, lines 32—33. We erroneously assumed a dynamical tropopause was computed. As a matter of fact a thermal definition is used within the model, compliant with the WMO definition.

page 28 Figure A2:

Please extent the y-Axis to accommodate all data points and provide information on the color code. Does the black line mean that you used only a Zmin(Φ) for your model degradation and not a Zmin(Φ,λ,t) as described on page 18? To me it seems that there is some seasonality. Would your analysis improve if you used a Zmin(Φ,t)? At high and very low latitudes (0-10N, 50-90N) the minimum altitude threshold seems to be below the median of the data points. What is your reason not using the median?

The y-axis was extended and some comment on the colour code was added to the legend.

To provide a slight correction: the black line is not Z_{min}, but Δ. For simplicity reasons, we did not consider any time dependency, and we deem the results obtained fair enough using this assumption.

As for the possible use of a mean or a median: our main goal was a demonstration of model degradation as a better approach to compare to observations, without going into an unnecessary detail of pixel-by-pixel correction. Therefore, though the derivation of Δ could of course have been carried out linearly regressing the OSIRIS data, we chose a qualitative approach, showing the first order effect. Furthermore, we were notified by personal communication (Adam Bourassa, Univ. Saskatchewan), that the OSIRIS sets of data were most likely to be updated in a near future, making the relevance of older versions of the data quite relative.

page 18 figure 7:

I suggest checking seasonality for your degradation. Unfortunately I cannot tell from Fig. A2 in which months your degradation altitude fits best, but at high latitudes you have a good agreement in October, November, April, and May and at low latitudes (0-20N) you have the yellow (day 250-350) and blue (day 425-525) features that might coincide with your data points above and below your Zmin(Φ). Please clarify.

As already stated, our degradation algorithm does not consider any time dependency. Due to the extended use of the Mie scattering code, it would have been cumbersome to check the effect of seasonality. This effect is possible, and likely, but was not investigated in the current study.

page 20 line 1-3:

I don't understand what you mean. Please detail where and to what extent the anomalies in Fig 8 agree better than the SAODs in Fig 7. Except from the shaded area indicating OSIRIS measurement gaps in the polar region in the middle panel of Fig. 8 I cannot see obvious additional information. Figure 7 already shows impressively that OSIRIS misses a substantial fraction of lower stratosphere sulfate aerosol.

The aim of calculating the anomalies is to set aside the background conditions, and to focus on the volcano effects only. We deem important to show that both the total SAODs and the anomaly optical depths match: the former is for comparison with previous studies, and the latter for the quantification of the volcanic effect.

page 20 figure 9:

Why are you showing 550 nm extinction here? It is not used anywhere else, all other OSIRIS data is presented for 750 nm. Please clarify and consider using less colors (7 might be sufficient) in the bottom figure. Some are indistinguishable.

550 nm is the standard output wavelength in CARMA; it is used here just for validation of the time evolution, and we chose to consider this readily exploitable output. Like for Fig. 1, we choose not to alter the number of colours, in order to keep a good rendition of the dynamics range.

page 21 line 3-5:

Please specify what you mean with "strongest measurement-biases shortly after the eruption". Do you mean OSIRIS high Zmin, or its saturation, or its rather coarse sampling that might miss local maxima of the plume filaments shortly after the eruption? Perhaps you want to compare with Günther et al. , ACPD (2017) Fig. 6, which is similar to your Fig. 9, but with different model and satellite data.

By "strongest measurement biases", we refer to the saturation process. This was clarified in the text. As for the comparison with Günther et al. (2017): our Fig. 9 is modelled extinction, whereas Günther et al.'s Fig 6 is modeled sulfur mass. Furthermore, the SO_2 mass is underestimated by MIPAS in first month.

page 21 table3, line 12-19:

For which purpose do you present e-folding times from other studies? They are not discussed here.

We present these values for quantification purposes. The noticeable result is the agreement between our degraded model and OSIRIS measurements.

The goal is not to discuss these in detail (they can depend firstly on many factors in the model, and also on how e-folding times are calculated, what period, etc.), but we do highlight that "One can note that the e-folding times vary quite significantly between authors, including those computed from OSIRIS's data (it is likely that different versions of the OSIRIS data —v.5.05 up to v.5.07 for the present study—were used)".

page 22 line 18/19:

Please add a reference for those removal processes.

We added the following reference:

Hamill, P., Jensen, E. J., Russell, P. B., & Bauman, J. J. (1997). The life cycle of stratospheric aerosol particles. *Bulletin of the American Meteorological Society*, *78*(7), 1395-1410.

page 22 line 25-28:

You might want to include the reff retrieval by Doeringer et al. (2012), who found 0.1—0.3 µm for the Sarychev, into your discussion.

This was added to the text.

"The latitudinal trend in reff simulated by our model is broadly consistent with the trend reported from ground-based remote sensing at Eureka (Nunavut, Canada) that found reff = 0.29 µm (O'Neill et al. (2012)), and with ACE measurements, which report reff = 0.1—0.3 µm (Doeringer et al. (2012)).

page 23 figure 10:

Please reduce number of colors. There are too many indistinguishable shades of red and green.

We choose to keep the number of colours as is, notably to maintain a good dynamic range.

page 22 line 34:

Which chlorine and bromine species do you mean?

They are ClO_x and BrO_x. This was added to the text in the revised version of the manuscript.

page 23 line 2:

Please note, the washout is not necessarily as efficient as in the Pinatubo case (von Glasow, Chemical Geology, 2009).

We agree with the referee. We add the clause "... though the washout for the Sarychev case was not necessarily as efficient [von Glasow, 2009]."

page 23 line 20 - page 24 line 2:

I did not understand this sentence. Please fix it.

We corrected the sentence to:

"This is primarily due to the higher aerosol loadings in these regions, and to favorable solar illumination conditions for which the catalytic ozone loss cycles (through OH radical production) are enhanced (Berthet et al., 2017)."

page 24 line 2-5:

I did not understand this sentence. Please reword and provide a reference.

We lightened the text, and replaced the concerned sentences with:

"A more detailed description of involved chemistry processes is provided in [Berthet et al., 2017]."

page 24 line 5:

I suggest starting a new paragraph here to clearly differentiate between heterogeneous reactions on aerosol particles and PSC particles. Isn't HCl the main reservoir of Cl and not ClONO2? There is HNO3 uptake by PSC particles that sediment out and hence lead to denitrification. I suggest to explicitly mention PSCs in this process and to reword this sentence.

As previously stated, the concerned paragraph was replaced.

It is not the scope of this paper to describe stratospheric chemistry linked to PSC.

page 24 line 16:

is 5% versus 7% a and 50 versus 60% a significant difference in your model? What is the uncertainty?

We cannot properly speak of uncertainty, since the model runs with a deterministic calculation (a large model ensemble study is beyond the scope of our work).

page 25 line 8:

I did not find your results convincing that an injection altitude of 11 to 15 km is realistic. I'd rather interpret your results that there are discrepancies between simulations and measurements above 15 km (see Fig. 4). Further your lower SAOD in the degraded model data compared to OSIRIS (Fig. 7 and 8) may be a result of not accounting for the SO2 injections into altitudes above 15 km that have been observed by several independent measurements.

We removed the clause "appears to be realistic", which we agree was too strong an assertion.

We added the following sentence: "The lack of more resolved data might be a source of uncertainty on the injection altitudes."

page 25 line 9-11:

Comparing your plume simulation in Fig 1 to IASI data and to the model simulations and AIRS data in Wu et al., ACP (2017), I find some shortcomings in this approach, which become also visible in several details and interpretations on which I commented before. Hence, I suggest to rephrase this sentence and add some discussion on potential errors due to the injection assumption.

We propose the following text change:

"The lack of more resolved data might be a source of uncertainty on the injection altitudes. However, this overall quantitative agreement reflects the model performance in SO_2 oxidation, atmospheric dispersion and aerosol processing. It indicates a suitable choice of eruption source parameters as used in previous studies e.g. Haywood et al. (2010) (an injection altitude ranging from 11 km to 15 km for SO_2, a vertical even spread of the total mass of gases injected, and a sole injection of the total gas mass on 15 June 2009, neglecting other minor injections on other days). These eruption source parameters did provide good results. They might need to be refined for model studies at higher temporal or spatial resolution, see Wu et al. (2017), Gűnther et al. (2017). We point out that an injected mass of 0.9 Tg SO2 (Clarisse et al., 2012; Realmuto and Berk, 2016) instead of 1.2 Tg of previous studies e.g. Haywood et al. (2010) is a fair hypothesis, and enables the model to closely reproduce the observed SO_2 burden according to the IASI retrievals of Clarisse et al. (2012)."

page 26 line 29-31: In which respect is this statement different from the findings in Ridley et al. (2014)?

We propose to keep this sentence as it is, because the findings of Ridley et al. (2014) (and similarly Mills et al., 2016) are more general in terms of temporal scale and volcanoes, are not directly related to OSIRIS SAOD, and only consider minimum altitude not the saturation effects in observation biases. Our study is focused on Sarychev eruption specifically, and presents a degradation approach to the model output (adjusted) to better compare to OSIRIS measurements.

We keep the sentence: "Our study therefore highlights that previous modelling studies (involving assumptions on particle size) that reported agreement to (biased) post-eruption estimates of SAOD derived from OSIRIS likely underestimated the climate impact of the 2009 Sarychev Peak eruption." but we propose to insert in the Introduction, page 3, line 32:

"More generally, underestimation of SAOD due to neglect of lower stratospheric volcanic aerosols has also been highlighted by Kravitz et al. (2011), Ridley et al. (2014), Andersson et al. (2015), Mills et al. (2016). As model studies of Sarychev eruption to date…"

Technical Suggestions

- *page 1, line 19: confirm*

This was corrected.

- *page 5 line 5: please sort references chronologically*

This was corrected.

- *page 5 line 7, 8: references for Sindelarova and Kettle are missing*

The concerned references were added to the reference list.

- *page 6 line 22: just write IASI*

This was corrected.

- *page 6 line 31: Define abbreviation at first usage only. Please also check in other places e.g. for OSIRIS page 7 line 20.*

These were corrected.

- *page 7 line 5: What does StraPolEte stand for? Is there any reference to the AEROWAVE" project?*

The acronym for StraPolÉte was already defined in the text.

We unfortunately have no paper reference for the AEROWAVE project.

- *page 10 figure 2: In the figure the IASI retrieval is green, but the caption says light blue.*

This was corrected.

- *page 11 line 10: "... Section 3.6." Please start a new paragraph here.*

A new paragraph was started there.

- *page 13 figure 4 caption: "solid" instead of "full" line*

This was corrected.

- *page 14 line 7: "... observations." Please start a new paragraph here.*

A new paragraph was started there.

- *page 14 line 12: Do the profiles only appear to be close to each other or are they close?*

They are close. This was corrected in the text.

- *page 18 line 15: Do you mean: A comparison for these months is therefore impossible?*

We replaced "difficult" by "not possible".

- *page 21 line 7: (9) = (Fig. 9)?*

This was corrected.

- *page 21 line 21/22: Please replace "... elsewhere, notably ..." by e.g. and refer to Table 3.*

This was corrected, and a reference to Table 3 was added.

- *page 22 line 17: ... , which*

This was corrected.

- *page 22 line 22: material*

This was corrected.

- *page 22 line 29: ...are thus a major ...*

This was corrected.

- *page 22 line 33: "... investigating the impacts of modern day eruptions on stratospheric..." Please fix this sentence.*

We rewrote the sentence as: "Most studies investigating the impacts of modern day eruptions on stratospheric chemistry..."

- *page 24 line 30: Please write "... over one day of eruption ..."*

This was corrected.

- *page 25 line 5: Please write "... suggest that the effective radius becomes ..."*

This was corrected.

All page and line numbers refer to the corrected version.

p. 1, l. 20: "confirms" was corrected to "confirm";

p. 2, l. 16: "global visible AOD was enhanced by up to 0.15" was changed to "global AOD (in the visible) was enhanced, reaching up to 0.15";

p. 2, l. 20: "a larger Antarctic ozone hole" was changed to "larger polar ozone holes". Reference to Tilmes et al. , 2008 was added.

p. 2, l. 21—27: The first part of the paragraph was rewritten as: "Moderate-magnitude explosive volcanic eruptions may also reach the stratosphere. However, they typically have a much reduced effect on climate and atmospheric chemistry compared to large-magnitude eruptions (Oman et al., 2005; Kravitz et al., 2010). In general a smaller mass of SO2 is injected and oxidized to sulfate aerosol. Also, by injecting to lower altitudes, the emissions from moderate-magnitude eruptions are more susceptible to removal by stratospheric-tropospheric exchange processes (Haywood et al., 2010)."

p. 2, l. 32: Mention to ash was added: "injecting SO2, ash and also Hcl...";

p. 3, l. 12: References were rewritten chronologically;

p. 3, l. 16: The following sentence was added to conclude the paragraph: "Further evidence for a larger particle size comes from effective radius estimate of 0.1—0.3 µm derived from satellite-based observations one month after the eruption (Doeringer et al., 2012) and a particle "sedimentation radius" of 0.5—1 µm from a model sensitivity study (Günther et al., 2017)."

p. 3, l. 33: The following sentence was inserted: "... lovermost stratosphere). More generally, underestimation of SAOD due to neglect of lower stratospheric volcanic aerosols has also been highlighted by Kravitz et al. (2011), Ridley et al. (2014), Andersson et al. (2015), Mills et al. (2016).

p. 5, l. 10: References were sorted out chronologically.

p. 5, l. 22: The following sentences were added: "The vertical distribution of our SO2 injection follows previous model studies. It is somewhat coarse approximation given that O'Neill et al. (2012) report lidar observations of fine-scale aerosol layers shortly after the eruption. Nevertheless, these were subsequently observed to collapse into a single layer in the lower stratosphere. For the magnitude of the SO2 injection we use a revised estimate that contrasts to previous studies, as discussed below."

p. 6, l. 29: "the IASI satellite instrument" was replaced with "IASI".

p. 7, l.2—5: The following sentences were inserted: "For this comparison we use the IASI retrieval of SO2 by Clarisse et al. (2012). The IASI dataset and retrieval algorithm used for this precise eruption can be considered as showing a lower threshold of around 0.3 DU, and SO2 loads can be expected to have a 10—20 % uncertainty. IASI retrievals have a typical altitude sensitivity of 1—2 km."

p. 7: "(Stratospheric and Tropospheric Aerosol Counter)" was moved to the first occurrence of "STAC", on line 4.

p. 7, l. 25: We added the sentence: “Note that both STAC and University of Wyoming OPCs have been compared by Renard et al. (2002).”

p. 7, l. 27—28: The sentence was reworked as: “Model SAOD was compared to that derived from extinction measurements by the OSIRIS (Optical Spectrograph and InfraRed Imaging System) aerosol instrument (onboard Odin satellite). OSIRIS is...”

p. 8, l. 11—12: The sentence was rewritten as: “There are some notable discrepancies for instance on 16 June 2009 south-west of Alaska: this is likely due to our simulation not accounting for the small amount of SO2 that was emitted before the main eruption on 15 June 2009.”

p. 8, l. 24—25: The following sentence was added: “The spatial and temporal evolution of the plume in our study is consistent with the results of Wu et al. (2017), where AIRS data are presented along with results of simulations by a particle dispersion model.”

Fig. 2, legend: “light blue” was corrected to “green”.

p. 8, l. 27 to p. 10, l. 4: The text was rewritten as: “Fig. 2 shows the modelled northern hemispheric SO2 burden in Tg, calculated by integrating the model anomalies from CESM1(WACCM) simulations with SO2 injection only and with SO2 and HCl co-injection (anomaly denotes a “volcano-on” simulation from which the “volcano-off” control run has been subtracted). Two adjusted CESM1(WACCM) model results are also presented that only include data over columns with > 0.3 DU SO2 to enable a better comparison to the IASI observations. Alongside is shown the observed evolution in northern hemispheric SO2 burden derived from the IASI retrieval by Clarisse et al. (2012) (that has a lower threshold of around 0.3 DU, see Methods 2.2). Finally, we also show the northern hemispheric SO2 burden as simulated using the HadGEM2 model (Haywood et al., 2010), and the IASI retrieval reported in that same study, both of which estimated 1.2 Tg SO2 injection in contrast to the revised IASI analysis (Clarisse et al., 2012) that yielded 0.9 Tg SO2 used in our study.”

p. 11, l. 6—14: The text was rewritten as: “A second notable result is that all the unadjusted model outputs overestimate the SO2 burden following the eruption compared to IASI measurements. The HadGEM2 model SO2 exceeds the Haywood et al. (2010) IASI observations for the whole period. The unadjusted CESM1(WACCM) outputs also exceed the Clarisse et al. (2012) IASI observations after a few days. This behaviour contrasts with the two adjusted CESM1(WACCM) model outputs that correct for the 0.3 DU SO2 lower value of the particular IASI retrievals used. The adjusted CESM1(WACCM) model outputs remain in close agreement to the observed post-eruption SO2 burden for the first 1–2 weeks, after which the model-simulated SO2 burdens decline more rapidly than the IASI 2012 observations. This evolution can be expected: a greater dispersion in the 22 model grid cells than in reality (and than observed by the IASI footprint of tens of kilometres), would cause an underestimation of the model SO2 burden compared to IASI. This effect will become more pertinent with dilution over time as the SO2 column approaches the 0:3 DU limit.”

Table 2: Decimal zeros were added, and in the text (“12.0” and “9.0”, p. 9, l. 9 & 12).

p. 12, l. 1—13: The text was reorganised as follows: “Our model-observation comparison of SO2 burden trends can also be quantified in terms of the e-folding time. The definition of the e-folding time is the following: let M(t) be the concentration of a species through time; if we assume it follows an exponential decay over a certain period of time t > t0, then is such as 8t > t0;M(t+) =M(t)=e, id est, corresponds to the time by which the concentration falls to 1=e of its initial value. For these calculations we choose the SO2 burden maximum as the initial value (0.9 Tg in our study). The e-folding time-constant for SO2 is approximately 17.0 days for the simulation including HCl, about two days longer than the approximately 15.0 days for the simulation that was run without HCl. When these CESM1(WACCM) model outputs are adjusted to correct for the 0.3 DU SO2 lower value of the particular IASI retrievals used they yield e-folding time-constants of 11.5 and 10.0 days, respectively. For the IASI SO2 retrieval of Clarisse et al. (2012) we calculate 12.0 days, i.e. very similar to the adjusted model simulation with SO2 and HCl co-injection (11.5 days). For comparison, Haywood et al. (2010) report that the HadGEM2 model yields a 13–14-day SO2 e-folding time (assuming a higher SO2 injection of 1.2 Tg and no HCl co-injection). Regarding IASI observations, Haywood et al. (2010) report an IASI SO2 e-folding time of 10–11 days, whilst using our method we calculate 9.0 days for the IASI retrieval of 2010. This is summarised in Table 2."

Fig. 4: Error bars were added on the dark and light blue curves.
In the legend, "full line" was replaced by "solid line".

p. 14, l. 23—25: The sentence was corrected to "The profiles from both volcano-on and volcano-off simulations are very close in the d > 20 nm size range (CN) indicating the progressive return of the simulated concentrations to background conditions for this size range."

p. 15, l. 15: The sentence was changed to: "This indicates the result of coagulation, condensation, sedimentation, and transport and dilution processes"

Fig. 5, legend: The following sentence was inserted: "Error bars on the total STAC counts can be evaluated to be +/-6%".

p. 18, l. 18—22: The text was corrected as: "These were chosen as a trade-off between the histogram of values in Fromm et al. (2014), and actual minimum altitudes reached by OSIRIS over the 2009–2010 period (see supplemementary material Fig. A2). For this series of calculations, thermal tropopause heights were diagnosed in the model."

Fig. 6 was reworked, changing the axes for some of the graphs.

Fig. 6, legend: The following sentence was inserted: "The measurement error on STAC measurements can be evaluated to be +/6%".

p. 18, l. 31: The sentence was changed to "A precise comparison for these months is therefore not possible."

p. 20, l. 11: We added the clause "due to the saturation effect".

p. 22, l.6—7: The sentence was rewritten as: "Discrepancies in the magnitude and e-folding times between model and OSIRIS SAOD's have been previously mentioned, e.g. Haywood et al. (2010); Kravitz et al. (2011); O'Neill et al. (2012), and are summarised here in Table 3."

p. 23, l. 5: We added a reference to Hamill et al., 1997.

p. 23, l. 13: We added the clause: "and with ACE measurements, which report reff = 0:1□0:3 m (Doeringer et al., 2012).

p. 23, l. 22—23: We added the clause: "though the washout for the Sarychev case was not necessarily as efficient (von Glasow et al., 2009)."

p. 23, l. 31-33: The sentences were changed to: "This is primarily due to the higher aerosol loadings in these regions, and to favorable solar illumination conditions for which the catalytic ozone loss cycles (through OH radical production) are enhanced (Berthet et al., 2017). A more detailed description of the involved chemical processes is provided in Berthet et al. (2017)."

p. 26, l. 8—14: The sentences were reworked as: "The lack of more resolved data might be a source of uncertainty on the injection altitudes. However, this overall quantitative agreement reflects the model performance in SO2 oxidation, atmospheric dispersion and aerosol processing. It indicates a suitable choice of eruption source parameters as used in previous studies e.g. Haywood et al. (2010) (an injection altitude ranging from 11 km to 15 km for SO2, a vertical even spread of the total mass of gases injected, and a sole injection of the total gas mass on 15 June 2009, neglecting other minor injections on other days). These eruption source parameters did provide good results. They might need to be refined for model studies at higher temporal or spatial resolution, see Wu et al. (2017), Günther et al. (2017)."

Fig. A2: A color code was added to the graph.

Model simulations of the chemical and aerosol microphysical evolution of the Sarychev Peak 2009 eruption cloud compared to in-situ and satellite observations

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Abstract. Volcanic eruptions impact climate through the injection of sulfur dioxide (SO_2), which is oxidized to form sulfuric acid aerosol particles that can enhance the stratospheric aerosol optical depth (SAOD). Besides large-magnitude eruptions, moderate-magnitude eruptions such as Kasatochi in 2008 and Sarychev Peak in 2009 can have a significant impact on stratospheric aerosol and hence climate. However, uncertainties remain in quantifying the atmospheric and climatic impacts of the 2009 Sarychev Peak eruption due to limitations in previous model representations of volcanic aerosol microphysics and particle size, whilst biases have been identified in satellite estimates of post-eruption SAOD. In addition, the 2009 Sarychev Peak eruption co-injected hydrogen chloride (HCl) alongside SO_2, whose potential stratospheric chemistry impacts have not been investigated to date. We present a study of the stratospheric SO_2-particle-HCl processing and impacts following Sarychev Peak eruption, using the CESM1(WACCM)-CARMA sectional aerosol microphysics model (with no a priori assumption on particle size). The Sarychev Peak 2009 eruption injected 0.9 Tg of SO_2 into the upper troposphere and lower stratosphere (UTLS), enhancing the aerosol load in the Northern hemisphere. The post-eruption evolution of the volcanic SO_2 in space and time are well reproduced by the model when compared to IASI (Infrared Atmospheric Sounding Interferometer) satellite data. Co-injection of 27 Gg HCl causes a lengthening of the SO_2 lifetime and a slight delay in the formation of aerosols, and acts to enhance the destruction of stratospheric ozone and mono-nitrogen oxides (NO_x) compared to the simulation with volcanic SO_2 only. We therefore highlight the need to account for volcanic halogen chemistry when simulating the impact of eruptions such as Sarychev on stratospheric chemistry. The model-simulated evolution of effective radius (r_{eff}), reflects new particle formation followed by particle growth that enhances r_{eff} to reach up to 0.2 µm on zonal average. Comparisons of the model-simulated particle number and size-distributions to balloon-borne in-situ stratospheric observations over Kiruna, Sweden, in August and September 2009, and over Laramie, U.S.A., in June and November 2009 show good agreement and quantitatively ~~confirms~~ confirm the post-eruption particle enhancement. We show that the model-simulated SAOD is consistent with that derived from OSIRIS (Optical Spectrograph and InfraRed Imager System) when both the saturation bias of OSIRIS and the fact

that extinction profiles may terminate well above the tropopause are taken into account. Previous modelling studies (involving assumptions on particle size) that reported agreement to (biased) post-eruption estimates of SAOD derived from OSIRIS likely underestimated the climate impact of the 2009 Sarychev Peak eruption.

1 Introduction

Explosive volcanic eruptions inject large quantities of sulfur dioxide (SO_2) into the atmosphere and have the potential to affect global climate (McCormick et al., 1995; Robock, 2000). Volcanic eruptions impact the global radiative budget via the formation of sulfuric acid aerosol particles from the volcanic SO_2 emitted. The presence of this particle load at stratospheric altitudes enhances the stratospheric aerosol optical depth (SAOD) and increases the solar backscatter, thereby inducing a cooling at the Earth's surface. The lifetime of sulfuric acid aerosol particles in the stratosphere can reach several years, significantly longer than in the troposphere (days to weeks). Large-magnitude eruptions that inject SO_2 directly into the stratosphere therefore typically have more prolonged and widespread (global or hemispheric) impacts than small-magnitude eruptions that typically inject SO_2 into the troposphere only. The June 1991 eruption of Mount Pinatubo was a large-magnitude eruption, with a Volcanic Explosivity Index (VEI, as defined in Newhall and Self (1982)) of 6, that had a significant impact on the stratospheric aerosol layer and hence climate (Bluth et al., 1992; Sato et al., 1993; Ammann et al., 2003): global ~~visible AOD was enhanced by~~ AOD (in the visible) was enhanced, reaching up to 0.15, causing a surface cooling of up to 0.5 degrees Celsius (Douglass and Knox, 2005; Wunderlich and Mitchell, 2017). In addition, stratospheric halogens (bromine and chlorine, that are present at elevated post-industrial concentrations in the stratosphere as consequence of past anthropogenic chlorofluorocarbon (CFC) emissions) became activated through reactions on the volcanic aerosol, causing substantial depletion of stratospheric ozone and ~~a larger Antarctic ozone hole (Portmann et al., 1996; Solomon et al., 1996)~~larger polar ozone holes (Portmann et al., 1996; Solomon et al., 1996; Tilmes et al., 2008).

Moderate-magnitude explosive volcanic eruptions may also reach the stratosphere~~, especially at high latitudes, where the tropopause height is lower~~. However, they typically have a much-reduced effect on climate and atmospheric chemistry compared to large-magnitude eruptions ~~(Oman et al., 2005; Kravitz et al., 2010). This is for several reasons. First, in~~ (Oman et al., 2005; Kravitz In general a smaller mass of SO_2 is injected and oxidized to sulfate aerosol. ~~Second~~Also, by injecting to lower altitudes, the emissions from moderate-magnitude eruptions are more susceptible to removal by stratospheric-tropospheric exchange processes ~~. Third, the impacts from moderate-magnitude eruptions at high-latitudes tend to be limited to one hemisphere only, in contrast to large eruptions at the tropics that inject and aerosol into both hemispheres.~~ (Haywood et al., 2010). Nevertheless, as volcanic eruption frequency follows an inverse power law with magnitude (e.g. Sparks (2003) and references therein), the cumulative impacts of frequent moderate-magnitude eruptions on stratospheric aerosol can be significant (Vernier et al., 2011), and for example were identified as a factor in recent decadal climate trends (Solomon et al., 2011; Ridley et al., 2014).

Here we study the moderate-magnitude eruption of Sarychev Peak volcano, which erupted in mid-June 2009 in the Kuril Islands, Russia (48°N; 153°E; 1512 m a.s.l.), injecting SO_2, ash, and also HCl to the stratosphere. The eruption was classified with a VEI of 4 (the volcanic eruptive index as defined in Newhall and Self (1982) is a logarithmic scale based on the volume of tephra ejected), and the main volcanic emission was injected to heights around 9–14 km as estimated from IASI retrievals (Carn et al., 2016; Carboni et al., 2016). Remote sensing observations over Eureka, Canadian Arctic, showed volcanic aerosol layers from the tropopause up to 16–17 km one month after the eruption, that subsequently settled into a more homogeneous layer in the lower stratosphere (O'Neill et al., 2012).

Previous modelling studies of the Sarychev 2009 eruption focused on the injection of SO_2, formation of volcanic aerosol and its radiative and atmospheric chemistry impacts (Haywood et al., 2010; Kravitz et al., 2011; Berthet et al., 2017). However, the models did not explicitly simulate the aerosol microphysical evolution of the volcanic cloud, rather they used bulk aerosol schemes and/or assumed size distributions. Model-simulated atmospheric impacts of the eruption on, for instance, aerosol optical depth, are highly dependent on the prescribed aerosol size (or effective radius, r_{eff}). An r_{eff} of $0.13\ \mu m$ was assumed in the HadGEM2 model study by Haywood et al. (2010), whilst Kravitz et al. (2011) used scaling to adjust their ModelE (Schmidt et al., 2006) simulations to represent a similar r_{eff}. Measurements that locally quantified the post-eruption volcanic aerosol include ground-based remote sensing ~~(Haywood et al., 2010; Kravitz et al., 2011; Mattis et al., 2010; O'Neill et al., 2012)~~ (Haywood et al., 2 balloon-borne observations. For example Kravitz et al. (2010) and Jégou et al. (2013) present in-situ balloon-borne observations of size-resolved stratospheric aerosol over Laramie, Wyoming, U.S.A. (June and November, 2009) and Kiruna, Sweden (August-September, 2009), respectively. The estimates of r_{eff} from all these measurements range from $0.1\ \mu m$ to $0.3\ \mu m$, i.e. larger than assumed in the models. Further evidence for a larger particle size comes from effective radius estimate of $0.1-0.3\ \mu m$ derived from satellite-based observations one month after the eruption (Doeringer et al., 2012) and a particle "sedimentation radius" of $0.5-1\ \mu m$ from a model sensitivity study (Günther et al., 2017).

O'Neill et al. (2012) highlight that this discrepancy can translate into large uncertainties in the modelled impacts, e.g. doubling of particle size from that assumed by Haywood et al. (2010) would lead to five-fold increase in the hemispherical (per particle) backscattering cross section of sulfate particles.

Volcanic aerosol from the Sarychev eruption also affected stratospheric halogen chemistry, via heterogeneous reactions on the aerosol surface area. The impacts were more modest than found for large-magnitude eruptions such as 1991 Mt. Pinatubo, but simulations suggest ozone depletion up to 4% in the lower stratosphere at high latitudes, with local NO_2 depletion up to 40% (Berthet et al., 2017), consistent with balloon-based and satellite observations (Adams et al., 2017).

To evaluate and tune the models, studies to date have relied upon satellite data from the OSIRIS instrument (Optical Spectrograph and Infrared Imaging System) to provide a global estimation of aerosol optical depth. However, comparison between OSIRIS and the models found a ≈ 1 month discrepancy in the timing of the SAOD maximum following the eruption. This was attributed to be likely due to deficiencies in the model aerosol microphysics, specifically the absence of nucleation processes (Haywood et al., 2010; Jégou et al., 2013). In subsequent work, Fromm et al. (2014) identified that stratospheric AOD derived from OSIRIS under high aerosol loadings was likely underestimated following volcanic eruptions, due to a saturation effect and because the extinction profiles may terminate well above the tropopause (and therefore miss volcanic aerosol in the lowermost stratosphere). More generally, underestimation of SAOD due to neglect of lower stratospheric volcanic aerosols has also been highlighted by Kravitz et al. (2011), Ridley et al. (2014), Andersson et al. (2015), Mills et al. (2016). As model studies to date have used OSIRIS-derived AOD's to evaluate and justify choice of model aerosol parameters such as r_{eff} (Haywood et al., 2010; Kravitz et al., 2011) this finding invokes the need to re-examine the assumed volcanic aerosol properties in the models. Finally, there have also been recent advances in satellite observations of volcanic gases in the stratosphere. First, new retrievals now enable an improved estimation of SO_2 mass injected combined with estimates of plume height from IASI (Infrared Atmospheric Sounding Interferometer) on the MetOp-A satellite (Clarisse et al., 2012; Carboni et al., 2016). Second, recent analysis of satellite data from the Microwave Limb Sounder (MLS) onboard satellite AURA identifies that Sarychev volcano co-injected HCl alongside SO_2 to the stratosphere (Carn et al., 2016). The co-injection of volcanic halogens alongside SO_2 could modify the resulting atmospheric chemistry/aerosol processing and impacts. In light of these advances it is instructive to perform a new model-observation study of the Sarychev 2009 eruption and its stratospheric impacts that furthermore benefits from recently developed model capabilities to simulate aerosol microphysics and size evolution.

Here we present model simulations of stratospheric aerosol evolution and chemistry following the moderate-magnitude 2009 Sarychev eruption using the global Community Earth System Model (CESM1) (Marsh et al., 2013), with its Whole Atmosphere Community Climate Model (WACCM) module for the simulation of the atmosphere, along with the sectional CARMA module (Community Aerosol and Radiation Model for Atmosphere (Toon et al., 1988)) to simulate aerosol microphysics. The sectional scheme distributes particles according to their size over 30 size bins, enabling the evolution of the particle size distribution to be traced in detail with no a priori assumptions on particle size. This model with sectional aerosol was previously used by English et al. (2013) to evaluate aerosol evolution and multi-year impacts from the large magnitude eruptions of Pinatubo 1991 and the $100\times$ larger Toba eruption (74000 years before present). Aerosol impacts from large-magnitude eruptions are substantial but limited by particle growth and sedimentation (with a 20-fold increase in AOD following Toba compared to Pinatubo despite its 100-fold increase in SO_2 injection). The globally averaged effective radius reached $0.45\ \mu m$ and $1.9\ \mu m$ after the Pinatubo and Toba eruptions, respectively. English et al. (2013) highlight the need to simulate microphysical processes and advantages of a sectional aerosol representation for a more comprehensive understanding of aerosol evolution following volcanic eruptions. This motivates our study that applies a sectional aerosol microphysics modelling approach to simulate aerosol evolution following a moderate-magnitude eruption.

The aims of our study are: i) to simulate the stratospheric aerosol evolution following the 2009 Sarychev eruption, using a model that explicitly accounts for aerosol microphysical processes using a sectional aerosol scheme. This will deliver the first model simulations of the size-resolved stratospheric aerosol evolution to assess impacts following the Sarychev eruption; ii) compare the model output to balloon-based in-situ measurements of size-resolved aerosol and to satellite observations of aerosol optical depth, including accounting for reported measurement limitations. This will deliver an improved model assessment of the aerosol impact in the 12 months following the Sarychev eruption; and iii) to investigate to what extent co-injection of HCl alongside SO_2 may have influenced the subsequent stratospheric aerosol processing and atmospheric chemistry impacts.

2 Methods

2.1 The CESM1(WACCM)-CARMA model: initialization, set-up and data post-processing

Model simulations were performed using the global Community Earth System Model (CESM1) using its Whole Atmosphere Community Climate module (WACCM) linked to the Community Aerosol and Radiation Model for Atmospheres (CARMA) module, involving the sulfur cycle with a sectional aerosol scheme (English et al., 2011). Land, sea-ice, and rivers were active modules, whereas oceans were data-prescribed. The spatial resolution was a longitude/latitude grid of 144 points by 96, respectively (i.e. approx. 2-degree resolution), and over 88 levels of altitude ranging from the ground to approximately 150 km altitude (with approx. 20 levels in the troposphere). Specified dynamics were used, with a nudging towards MERRA meteorological data (Rienecker et al., 2011) at every time step (30 min) with a weight factor of 0.1 towards the analysis, for temperature and wind fields. The following surface emissions were prescribed in the model. For SO_2, NH_3, black carbon, organic carbon, NO_x, CH_4 and CO emissions, the MACCity data set was used ~~(Granier et al., 2011; Diehl et al., 2012; Lamarque et al., 2010; van der Werf et al.,~~ Anthropogenic CH_4 emissions were added from the EDGAR v4.2 database (available at http://edgar.jrc.ec.europa.eu) biogenic CO emissions were added from the MEGAN-MACC database (Sindelarova et al., 2014). OCS was prescribed using data from Kettle et al. (2002). CH_2O was input according to the IPCC RCP8.5 scenario (Riahi et al., 2011), and for H_2 the ECCAD-GFED3 database was used (van der Werf et al., 2010). For CO_2, N_2O, CCl_4, CF_2ClBr, CF_3Br, CH_3Br, CH_3CCl_3, CH_3Cl, CFC11, CFC113, CFC12 and HCFC22 emissions, lower boundary conditions were prescribed following CCMI/RCP8.5 data.

Simulations were set to start on 1st January 2009, using the CESM1(WACCM) initial atmosphere state file at that date. This enabled a six-month model spin-up period before the eruption injection on 15th June 2009, after which the simulations were continued for one year, ending on 31st May 2010. The Sarychev Peak eruption was simulated by injecting volcanic SO_2 (and HCl) gases into model grid boxes corresponding to the location of the volcano (48°N;153°E), over the duration of 15 June 2009, spread evenly between 11 km and 15 km altitude a.s.l. The model's $2.5°$ longitude $\times$ $1.875°$ latitude grid resolution means that the volcanic plume is initially too dilute in the model compared to reality. This is nevertheless common methodology, see e.g. Haywood et al. (2010). The vertical distribution of our SO_2 injection follows previous model studies. It is a somewhat coarse approximation given that O'Neill et al. (2012) report lidar observations of fine-scale aerosol layers shortly after the eruption. Nevertheless, these were subsequently observed to collapse into a single layer in the lower stratosphere. For the magnitude of the SO_2 injection we use a revised estimate that contrasts to previous studies, as discussed below.

A detailed chronology of the Sarychev Peak 2009 eruption can be found in Levin et al. (2010) that identified three explosive periods: on 12-13 June, repeated explosions occurred, reaching heights ranging from 5 km to 10 km; an isolated, high-altitude explosion occurred on 14 June, reaching 21 km altitude; finally, on 15 June, a series of consecutive explosions reached altitudes ranging between 10 km and 15 km (all times are in UTC). The first eruptive clouds on the 11–14 June period were mainly ash (Rybin et al., 2011). We neglected the minor, low-altitude (inferior to 5 km) explosions reported on 11 and 16 June, and injected SO_2 continuously for a 24-hr period on 15 June spread evenly between 11 km and 15 km altitude a.s.l. The timing of the SO_2 emissions is based on SO_2 satellite retrievals from IASI (Clarisse et al., 2012; Carn et al., 2016; Carboni et al., 2016), MODIS (MODerate-resolution Imaging Spectroradiometer) (Rybin et al., 2011; Realmuto and Berk, 2016), and OMI (Ozone

Monitoring Instrument) (Theys et al., 2015) which all show that the majority of high altitude SO_2 was released on the 15^{th} (and possibly in the early morning of the 16^{th}). Haywood et al. (2010) used a total injection mass of 1.2 Tg SO_2, which was the SO_2 total mass value retrieved on 16 June with IASI. An update of the SO_2 algorithm (Clarisse et al., 2012) found a maximum SO_2 mass value of around 0.9 Tg; a value which was confirmed with subsequent updates of that algorithm (Carn et al., 2016). It is also consistent with retrievals from OMI (Theys et al., 2015) and MODIS (Realmuto and Berk, 2016). In contrast, the IASI retrievals reported in Carboni et al. (2016) found that the transient SO_2 burden reached only up to 0.6 Tg SO_2. We consider though that 0.9 Tg of SO_2 is the best estimate for the mass of SO_2 injected by Sarychev peak into the UTLS. We did not consider any ash emissions.

In a second simulation, 27 Gg HCl was co-injected alongside the 0.9 Tg of SO_2. This initialization follows the recent identification of a localized stratospheric HCl enhancement following the Sarychev eruption (Carn et al., 2016), based on analysis of Microwave Limb Sounder (MLS) satellite observations, reporting a HCl/SO_2 mass ratio of around 3%. Since the low vertical resolution of MLS in the lower stratosphere makes it difficult to infer the precise injection altitude of HCl, we assumed an HCl injection altitude identical to that of SO_2. A control run without the volcanic gas injection was also performed, enabling anomalies to be calculated. In the present paper, we will refer to control runs as "volcano-off" simulations, and to runs including the eruption as "volcano-on" simulations.

The CESM1(WACCM) atmospheric chemistry scheme includes a detailed sulfur cycle and key stratospheric nitrogen (NO_y), halogenated (i.e. chlorine and bromine) and hydrogenated (in particular HO_x radicals) compounds. The formation and microphysics of sulfuric acid aerosol particles simulated by the CARMA module is described in detail in English et al. (2011).

The CARMA module in sectional configuration yields particle concentration across 30 size-bins ranging from approximately 0.68 nm to 3.25 µm in dry diameter. Effective radius is also provided as a direct model output. Post-processing of the model output was used to determine wet particle size distributions, extinctions and optical depth. In each model grid-cell, the wet diameter of each size-bin was calculated using a (hygroscopic growth) parameterisation of $H_2SO_{4(aq)}$ particle volume as a function of acid weight percentage ($wt\%H_2SO_4$), ambient humidity and temperature following Tabazadeh et al. (1997). Extinctions at 750 nm and 550 nm were calculated by combining the particle concentrations across the sectional size bins with the corresponding wet radii and particle refractive indices following Beyer et al. (1996), using a Mie scattering code at the desired wavelength (van de Hulst and Twersky, 1957). The aerosol extinctions were integrated with altitude over the stratosphere to yield stratospheric aerosol optical depth (SAOD).

2.2 Balloon-borne in-situ and satellite-based remote sensing observations of aerosol and SO_2

The model SO_2 output (from simulations with and without HCl co-injection) is compared to vertical columns of SO_2 and total (northern hemispheric) SO_2 burden derived from ~~the IASIsatellite instrument~~IASI. The Infrared Atmospheric Sounding Interferometer is an instrument present on board the MetOp-A satellite since the end of 2006. It is a spectrometer measuring infrared light spectra at nadir. Its primary goal is to assess for temperature and water vapour content of the atmosphere, but it can also be used to retrieve the atmospheric concentrations of various gases, amongst which SO_2 (Clarisse et al., 2008; Carboni et al., 2016). IASI provides global coverage twice a day and its footprint ranges from circular (12 km diameter at nadir) to elliptical (up to 20 km by 39 km at the end of the swath). For this comparison we use the IASI retrieval of SO_2 by Clarisse et al. (2012). The IASI dataset and retrieval algorithm used for this precise eruption can be considered as showing a lower threshold of around 0.3 DU, and SO_2 loads can be expected to have a 10–20% uncertainty. IASI altitude retrievals have a typical sensitivity of $1-2$ km. We also compare our results to HadGEM2 model simulations of SO_2 and earlier IASI SO_2 retrievals reported by Haywood et al. (2010).

Comparisons of the modelled aerosols with in-situ measurements are two-fold. First, we compare the model's output with size-resolved aerosol measurements carried out with the balloon-borne STAC (Stratospheric and Tropospheric Aerosol Counter) Optical Particle Counter (OPC) instrument over Kiruna, Sweden, on 2, 7, 18 August 2009 and 18 May 2010. STAC ~~(Stratospheric and Tropospheric Aerosol Counter)~~ can be borne under stratospheric balloon gondolas, and can measure low concentrations in aerosols (down to approximately 10^{-4} cm^{-3}µm^{-1} (Ovarlez and Ovarlez, 1995; Renard et al., 2005, 2010). Particles are classified by their diameters into tuneable size-bins ranging from a few tenths of micrometre to a few micrometres. The counts in each size bin are normalised by the bin width to yield a size distribution. The uncertainty, defined as the relative standard deviation, is 60% for aerosol concentrations of 10^{-3} cm^{-3}, 20% for 10^{-2} cm^{-3}, and 6% for concentrations higher than 10^{-1} cm^{-3}. STAC was operated successfully on eight different balloon flights throughout the August–September 2009 period over Kiruna, Sweden (68°N; 20°E), as part of the StraPolÉté campaign (French acronym for Stratosphère Polaire en Été), and also in May 2010, as part of the AEROWAVE project (acronym for AEROsols, WAter Vapour and Electricity). Measurements of the STAC instruments are available online at http://www.pole-ether.fr. During these flights, it was demonstrated that STAC passed through the Sarychev plume (Jégou et al., 2013), as explored further in the present paper. Our comparison focuses on the submicron range between ≈0.3 µm and 1 µm diameter. We have performed an interpolation of the counts from the model's size bins to the STAC size bins (and from the model pressure levels to the observed pressure of the balloon payload) in order to enable a direct comparison.

Second, we also compare the model's aerosol output with in-situ measurements carried out by the OPC of the University of Wyoming (Deshler et al., 2003), flown on stratospheric balloons launched from Laramie, U.S.A. (41°N; 105°W), on 22 June 2009 and 7 November 2009 (Kravitz et al., 2011). For comparison to the model, total particle number above two diameter threshold sizes are considered here: $d > 20$ nm (condensation nucleii, CN) and $d > 0.5$ µm (N). Uncertainties are 85%, 25% and 8% for concentrations of 10^{-3} cm^{-3}, 10^{-2} cm^{-3} and 10^{-1} cm^{-3} respectively (Deshler et al., 2003). These data are available from ftp://cat.uwyo.edu/pub/permanent/balloon/Aerosol_InSitu_Meas/US_Laramie_41N_105W/. They have been derived by the University of Wyoming as follows: the measurement of N is calculated directly from the OPC instrument. The CN is derived from a condensation nuclei counter co-deployed on the balloon payload. Note that both STAC and University of Wyoming OPCs have been compared by Renard et al. (2002).

Model SAOD was compared to that derived from extinction measurements by the OSIRIS ~~aerosol instrument (onboard Odin satellite). The~~ (Optical Spectrograph and InfraRed Imaging System~~(OSIRIS)~~) aerosol instrument (onboard Odin satellite). OSIRIS is a limb sounder able to provide information on the vertical distribution of atmospheric aerosols (Bourassa et al., 2007, 2008) from the upper troposphere up to the lower mesosphere through the analysis of scattered sunlight. This Canadian instrument has been active since November 2001 on board Swedish satellite Odin (Llewellyn et al., 2004). Its global coverage reaches up to 82° in latitude. Odin evolves on a sun-synchronous orbit, and therefore the availability of OSIRIS's measurements is latitude- and time- dependent. Our analysis focuses on extinction measurements from OSIRIS version 5.07, available from http://odin-osiris.usask.ca/. Importantly, a novel aspect of our study is that our analysis specifically accounts for instrument errors or limitations as reported by Fromm et al. (2014). Model output data have been degraded accordingly. First, the modelled extinctions have been made to saturate at an upper threshold of 2.5×10^{-3} km^{-1}; then, extinctions have been only integrated above a certain altitude, dependent on the latitude: a linear variation of this lower limit was assumed, from 0.5 km above the tropopause at the equator up to 5.5 km above the tropopause at the poles. Further details are given in Results, Section 3.4.

3 Results

3.1 Spatial and temporal evolution of volcanic SO_2 vertical column densities

Fig. 1 shows vertical column densities of SO_2 from the CESM1(WACCM) simulation in which both volcanic SO_2 and HCl were injected (right-hand panel) and a comparison with IASI retrievals (left-hand panel). Both sets of maps are shown with the same lower threshold in terms of Dobson units, corresponding to an estimated lower threshold of 0.3 DU in IASI's retrievals for this precise eruption and for the IASI retrieval algorithm used (Clarisse et al., 2012). The spatial and temporal evolution of the Sarychev SO_2 plume is reasonably well simulated by the CESM1(WACCM) runs throughout the first fortnight following the eruption. There are some notable discrepancies for instance on 16 June 2009 south-west of Alaska: this is ~~because our simulation does not account~~ likely due to our simulation not accounting for the small amount of SO_2 that was emitted before the main eruption on 15 June 2009. Also, Asian pollution (close to 0.3 DU) is evident in the simulations shown in Fig. 1 but not observed by IASI, likely due to the reduced sensitivity of the IASI retrievals to SO_2 below 5 km altitude. To quantify the spatial-amplitude match between the two sets of data, we chose a colocation index calculated as:

$$\rho = \frac{\mathbb{E}[(P_1 - \mu_1)(P_2 - \mu_2)]}{\sigma_1 \sigma_2} \quad (1)$$

where P_1 and P_2 are the bi-dimensional matrices representing the spatial SO_2 loads (for model and satellite retrievals), sampled over the same spatial grid and stacked into monodimensional vectors; $\mu_{\{1;2\}}$ and $\sigma_{\{1;2\}}$ are their respective means and standard deviations. It is expected that the index drops quite quickly after the eruption due to greater dispersion in the model on the 2×2 degree grids than in the finer-scale (tens of km) IASI observations. Colocation indices were calculated over the first fortnight following the eruption, (Table 1), for the simulation with SO_2 injection only and with HCl co-injection alongside SO_2. As can be noted from Table 1, co-location indices show comparable values for both model runs. This indicates broadly similar SO_2 dispersion in the model runs.

The spatial and temporal evolution of the plume in our study is consistent with the results of Wu et al. (2017), where AIRS data are presented along with results of simulations by a particle dispersion model.

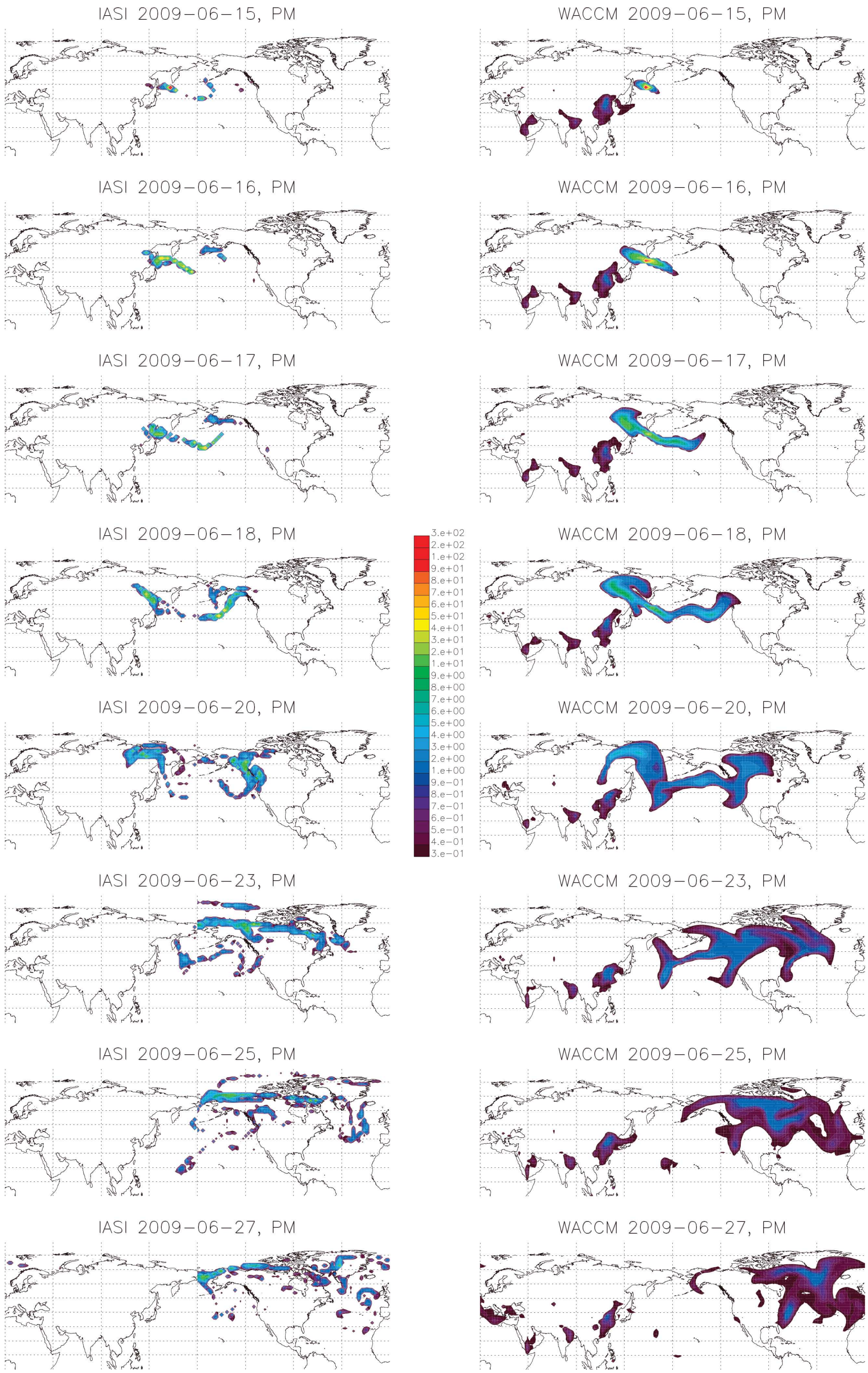

Figure 1. Spatial and temporal evolution of vertical column densities of SO_2 (in Dobson Units, DU) over 1–2 weeks following the Sarychev eruption according to IASI satellite observations (left) and simulated by the CESM1(WACCM) model (right). A threshold of 0.3 DU was applied, corresponding to the lower threshold for this precise IASI retrieval (Clarisse et al., 2012). The CESM1(WACCM) model data correspond to instantaneous output at midnight, whereas the IASI data are gathered over the whole of the post-meridiem period.

Date	15 June	16 June	17 June	18 June	20 June	23 June	25 June	27 June
Colocation index, with HCl, in %	90.34	41.29	17.61	20.26	16.60	12.38	11.36	5.36
Colocation index, without HCl, in %	90.36	41.25	17.55	20.23	16.62	12.85	11.30	5.48

Table 1. Colocation indices quantifying the spatial-amplitude agreement in the volcanic SO_2 vertical column densities simulated by CESM1(WACCM) compared to IASI observations over the northern hemisphere, for 1–2 weeks after the eruption (dates corresponding to Fig. 1). See Eq. 1 for details of the computation.

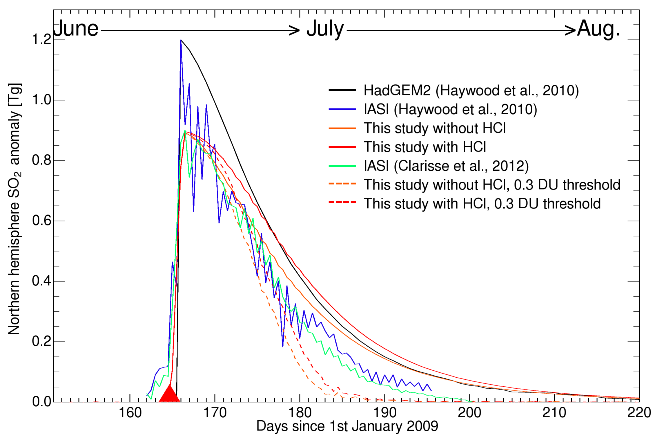

Figure 2. Temporal evolution of the SO_2 burden (in Tg), integrated over the Northern hemisphere over June–August 2009. Model anomalies are shown for simulations that injected SO_2 only (orange) and with co-injection of HCl (red), alongside the IASI retrieval (~~light blue~~green) with maximum burden of 0.9 Tg SO_2. Also shown are adjusted model outputs that account for the 0.3 DU IASI lower threshold for this particular case (red and orange dashed lines). For comparison, the previously reported model study and IASI retrieval of Haywood et al. (2010) that assumed a higher maximum burden of 1.2 Tg SO_2 are also depicted (black and blue lines, respectively).

3.2 Lifetime, burden of volcanic SO_2, and role of co-injected HCl

Fig. 2 shows the modelled northern hemispheric SO_2 burden in Tg, calculated by integrating the model anomalies from CESM1(WACCM) simulations with SO_2 injection only and with SO_2 and HCl co-injection (anomaly denotes a "volcano-on" simulation from which the "volcano-off" control run has been subtracted). ~~Alongside is shown the observed evolution in northern hemispheric burden derived from the IASI retrieval by Clarisse et al. (2012). The IASI set of data used for this precise eruption, and considering the retrieval algorithm used (Clarisse et al., 2012), can be considered as showing a lower threshold of around , therefore two~~ Two adjusted CESM1(WACCM) model results are also presented that only include data over columns with ~~$>$~~ $>$ 0.3 DU SO_2 to enable a better comparison to the IASI observations. Alongside is shown the observed evolution in northern hemispheric SO_2 burden derived from the IASI retrieval by Clarisse et al. (2012) (that has a lower threshold of around 0.3 DU, see Methods 2.2). Finally, we also show the northern hemispheric SO_2 burden as simulated using the HadGEM2 model

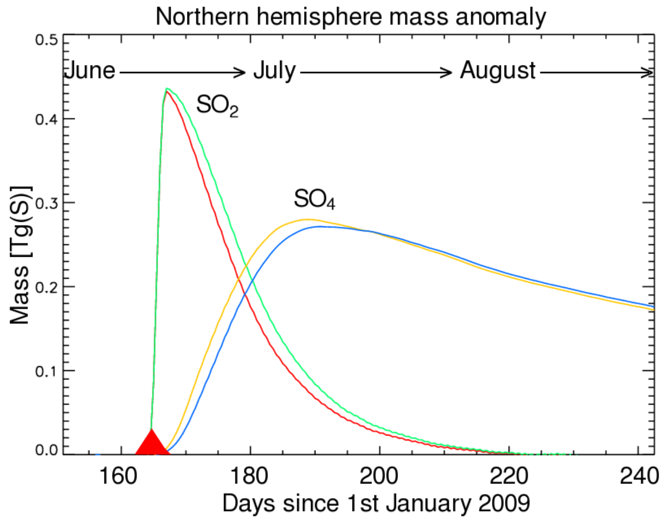

Figure 3. Temporal evolution of total SO_2 and SO_4 burdens (Tg sulfur), integrated over the Northern hemisphere over June–August 2009 (the eruption is depicted by the red triangle). Model anomalies are shown for runs with injection of SO_2 only (red and yellow for SO_2 and SO_4 respectively) and with co-injection of HCl (green and blue for SO_2 and SO_4 respectively).

(Haywood et al., 2010), and the IASI retrieval reported in that same study, both of which estimated 1.2 Tg SO_2 injection in contrast to the revised IASI analysis (Clarisse et al., 2012) that yielded 0.9 Tg SO_2 used in our study.

A notable result is the slower decline in SO_2 burden for the model run with volcanic SO_2 and HCl co-injection than volcanic SO_2 (only). There is also a corresponding slower increase in the sulfate aerosol burden (Fig. 3).

The presence of HCl slows down the oxidation of SO_2 to sulfuric acid aerosol particles and hence lengthens the e-folding time of SO_2 in the stratosphere by about two days (see calculations below). This is as a result of the competition between their two main oxidation reactions involving OH. These are:

$$HCl + OH \rightarrow Cl + H_2O \tag{R1}$$

and the trimolecular reaction (where M is a third-body, e.g. N_2 or O_2):

$$SO_2 + OH + M \rightarrow HSO_3 + M \tag{R2}$$

where HSO_3 subsequently leads to the formation of H_2SO_4 through the reaction sequence described by Weisenstein et al. (1997). This conversion of SO_2 to H_2SO_4 is limited by the rate of R2 below 40 km in altitude. Competition between R2 and R1 results in a slower rate of oxidation of volcanic SO_2 in the presence of co-injected HCl.

A second notable result is that all the ~~(unadjusted)~~ unadjusted model outputs overestimate the SO_2 burden following the eruption compared to IASI measurements. ~~Conversely the~~ The HadGEM2 model SO_2 exceeds the Haywood et al. (2010) IASI observations for the whole period. The unadjusted CESM1(WACCM) outputs also exceed the Clarisse et al. (2012) IASI observations after a few days. This behaviour contrasts with the two adjusted CESM1(WACCM) model outputs ~~(to~~ that correct

	This study, model with HCl	This study, model without HCl	Haywood et al. (2010) HadGEM2 model	Haywood et al. (2010) IASI 2010	This study, IASI 2010	This study IASI 201
SO_2 e-folding time	~~≈ 17~~≈ 17.0 days	~~≈ 15~~≈ 15.0 days	≈ 13 ≈ 14 days	N.A.	N.A.	N.A
With 0.3 DU threshold	≈ 11.5 days	~~≈ 10~~≈ 10.0 days	N.A.	≈ 10 ≈ 11 days	~~≈ 9~~≈ 9.0 days	~~≈ 12~~≈ 12.0

Table 2. Comparison of the calculated SO_2 e-folding times for this study and Haywood et al. (2010). Two sets of IASI data are investigated: 2010 and 2012 retrievals. For the sake of the comparison with the satellite data, a lower threshold of 0.3 DU is applied whenever possible. The most recent IASI data (2012) yield a value close to that calculated with the model simulation of this study with SO_2 and HCl co-injection.

for the 0.3 DU SO_2 lower value ~~for~~ of the particular IASI retrievals used~~)~~. The adjusted CESM1(WACCM) model outputs remain in close agreement to the observed post-eruption SO_2 burden for the first 1–2 weeks, after which the model-simulated SO_2 ~~burden declines~~ burdens decline more rapidly than the IASI 2012 observations~~(Clarisse et al., 2012)~~. This evolution can be expected: a greater dispersion in the ~~$2° \times 2°$~~ $2° \times 2°$ model grid cells than in reality (and than observed by the IASI footprint of tens of kilometres), would cause an underestimation of the model SO_2 burden compared to IASI. This effect will become more pertinent with dilution over time as the SO_2 column approaches the 0.3 DU limit.

In summary, we find that the CESM1(WACCM) model run (adjusted output) with SO_2 and HCl co-injection gives best agreement to the IASI SO_2 observations. The simulation with SO_2 with HCl injection therefore forms the basis for further analysis in Section 3.6.

Our model-observation comparison of SO_2 burden trends can also be quantified in terms of the e-folding time. The definition of the e-folding time τ is the following: let $M(t)$ be the concentration of a species through time; if we assume it follows an exponential decay over a certain period of time $t > t_0$, then τ is such as $\forall t > t_0, M(t+\tau) = M(t)/e$, id est, τ corresponds to the time by which the concentration falls to $1/e$ of its initial value. For these calculations we choose the SO_2 burden maximum as the initial value (0.9 Tg in our study). The e-folding time-constant for SO_2 is approximately ~~17~~17.0 days for the simulation including HCl, about two days longer than the approximately ~~15~~15.0 days for the simulation that was run without HCl. ~~For~~ When these CESM1(WACCM) model outputs are adjusted to correct for the 0.3 DU SO_2 lower value of the particular IASI retrievals used they yield e-folding time-constants of 11.5 and 10.0 days, respectively. For the IASI SO_2 retrieval of Clarisse et al. (2012) we calculate 12.0 days, i.e. very similar to the adjusted model simulation with SO_2 and HCl co-injection (11.5 days). For comparison, Haywood et al. (2010) report that the HadGEM2 model yields a 13–14-day SO_2 e-folding time (~~without the injection, and~~ assuming a higher SO_2 injection of 1.2 Tg and no HCl co-injection). Regarding IASI observations, Haywood et al. (2010) report an IASI SO_2 e-folding time of 10–11 days, whilst using our method we calculate ~~9~~9.0 days for the IASI retrieval of 2010. ~~For the IASI retrieval of Clarisse et al. (2012) we calculate 12 days, i.e. very similar to the adjusted model simulation with and co-injection (11.5 days).~~ This is summarised in Table 2.

3.3 Comparison of the model to in-situ balloon-based measurements of size-resolved aerosol

Here we compare size-resolved aerosol concentrations from our simulations with in situ measurements from balloon-borne OPCs over Laramie, USA (June, November, 2009) and Kiruna, Sweden (August, September, 2009). It should be emphasized that the instruments are likely to detect a wider range of particles and particle compositions present in the stratosphere, i.e. internally and externally mixed particles with some organic and meteoric component (Murphy et al., 2014), whereas our model simulations provide pure sulfuric acid aerosol particles only. Nevertheless, these are expected to be the dominant source of aerosol in the lower stratosphere in the months following the Sarychev Peak 2009 eruption.

First we compare the model to measurements carried out by the University of Wyoming OPC (Deshler et al., 2003) during balloon-borne flights over Laramie, Wyoming (U.S.A., $41°$N, $105°$W) on 22 June 2009 and 7 November 2009. These observations were made one week and nearly five months after the Sarychev eruption, respectively. Kravitz et al. (2011) previously suggested that a significant volcanic influence can be seen in the data from 7 November but not on 22 June, based on comparison with balloon flights from other years. Here we compare the data directly to aerosol simulated by our model runs.

Fig. 4 shows both the model and measured aerosol particle number concentrations over Laramie for two particulate size ranges: $d > 20$ nm (noted CN, for condensation nucleii) and $d > 0.5$ μm (noted N). Overall there is good general agreement between simulated and measured values in terms of number concentrations and in the general trend with respect to altitude and size range separation. Note that model-measurement differences are greater in the troposphere since only sulfuric acid particles are simulated.

The upper panel of Fig. 4 (22 June 2009) shows that the volcano-off simulation reproduces the in situ observations of particle number with a very good agreement, supporting the hypothesis of Kravitz et al. (2011) that there was no significant volcanic influence on this day. However, the volcano-on simulation in fact simulates the presence of a volcanic plume, as can be seen by enhancements in CN and N between 13 km and 15 km altitude. We note that the precise geographical location of plume structures is difficult to simulate using low resolution simulations just one week after the eruption. Remote sensing observations suggest the initial presence of multiple aerosol layers in the stratosphere that subsequently collapsed into a single layer (O'Neill et al., 2012), whereas our CESM1(WACCM) model study assumes injection over 11–15 km. We also suggest that model horizontal resolution effects are a further possible source of error in the volcano-on simulation that might have led to anomalous sulfate plume structure over the measurement location. A geographic 2-D map of the vicinity of Laramie that shows model-simulated sulfuric acid aerosol particles at 13 km altitude, Fig. A1, shows that the location of the measurements lies on the edge of an aerosol plume structure simulated by the model. Diffusion on the model grids ($2° \times 2°$ resolution) or uncertainties in the initialisation altitude could therefore lead to modelled plume structure over Laramie that is not evident in the observations.

Conversely, on 7 November the volcanic plume is simulated to be much more homogeneous (and dilute), covering a larger area that encompasses Laramie. The lower panel of Fig. 4 shows modelled and observed aerosol particle number concentrations for 7 November 2009. For the $d > 0.5$ μm size range (N), the agreement between the volcano-on simulation and the in-situ measurements below 17 km indicates that volcanic aerosol particles were still present and detectable over Laramie nearly five

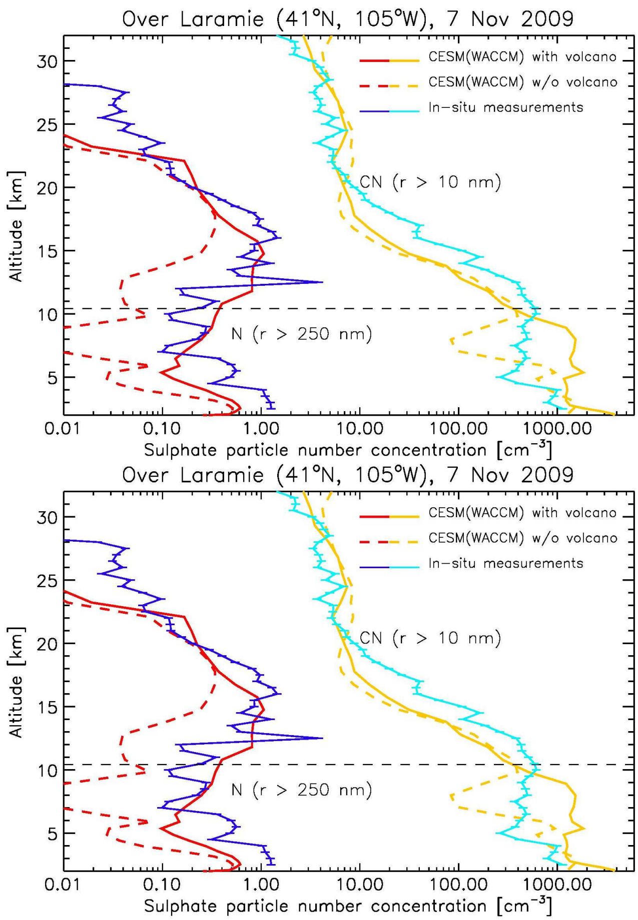

Figure 4. Comparison of particle number concentration over Laramie (U.S.A., 41°N, 105°W), simulated by the CESM1(WACCM) model (red/orange lines: simulations with and without the Sarychev eruption are shown as ~~full~~ solid and dashed lines, respectively) and by balloon-borne in situ measurements (blue/cyan lines), for 22 June 2009 and 7 November 2009. Two size ranges are shown: $d > 20$ nm (CN) and $d > 0.5$ µm (N). The model tropopause height is depicted by the dashed grey line. Model uncertainties are greater in the troposphere. On 22 June 2009, the presence of a volcanic plume over Laramie is simulated in the model (evident in both CN and N at the tropopause, discussed in the text), but not evident in the observations. On 7 November 2009 the presence of a more dilute volcanic plume is simulated in the model (evident in N only) that is consistent with the observed N in the lower stratosphere

months after the eruption, and their presence can be quantitatively reproduced by the CESM1(WACCM) model. The profiles from both volcano-on and volcano-off simulations appear are very close in the $d > 20$ nm size range (CN) indicating the progressive return of the simulated concentrations to background conditions for this size range.

Next we compare the CESM1(WACCM) simulations to in situ aerosol measurements made by the STAC instrument on a balloon gondola in northern Sweden. Fig. 5 compares the particle counts observed by STAC, and the sulfate particle concentrations simulated by the WACCM model for the same location (Kiruna, Sweden, 67°N, 20°E), and times: 2, 7 and 18 August 2009. A comparison is also shown for 18 May 2010 when the stratosphere can be considered to be close to background conditions. The model outputs have been interpolated to the pressure observed by the balloon payload, and to the specific size-bins of the STAC instrument covering 0.325 to 0.885 µm mean diameter.

In Fig. 5, a volcanic sulfate aerosol plume can clearly be identified between 11 km and 19 km altitude for all flights in August 2009. This is demonstrated in the third column of the figure by an important difference in modelled particle number over the size-bins of the STAC for the volcano-on and volcano-off simulations: total particle number on the STAC diameter range is enhanced by the volcanic eruption by between one and two orders of magnitude depending on the altitude. Total number simulated in the volcano-on simulation is in good general agreement to the STAC observations.

There are some discrepancies between model and observations at higher and lower altitudes: at lower altitudes, the model yields lower counts than the instrument's counts: this is likely due to the presence in the troposphere of non-sulfate aerosols unaccounted for by the model. For the discrepancies above the plume's altitude, the radiometer MicroRADIBAL (French acronym for Micro RADIomètre BALlon) (Brogniez et al., 2003; Renard et al., 2008), flown alongside STAC, identified the presence of some light-absorbing particles around 20 km altitude (Jégou et al., 2013): these might have affected the STAC measurements (STAC is designed for sulfate particle detection) and were also not included in the model. Their origin is still to be determined. Nevertheless, the good agreement in total number between model and observations in the lower stratosphere (corresponding to the main influence of the volcanic plume) confirms the strong impact of the Sarychev eruption on aerosol number.

Comparing these aerosol observations above Kiruna in August to those above Laramie in November on an order of magnitude basis, the Laramie measurements have ≈ 1 cm^{-3} particulates of diameter greater than 0.5 µm at 14 km altitude in November, whereas measurements over Sweden in August of the same year show approximately 10 to 100 times more particles of size greater than 0.4 µm in diameter. This indicates the result of coagulation, condensationand sedimentation, sedimentation, and transport and dilution processes: two months after the eruption there is a strong volcanic impact, but few sub-micrometer size volcanic particles are left in the stratosphere five months after the eruption. The volcanic aerosol evolution is discussed further in Section 3.5 below.

Fig. 6 shows the particle size distributions measured by the STAC, separated in 1-km layers of altitude, for the same four flights as Fig. 5, and compares these to size distributions simulated by the CESM1(WACCM) model. The size distributions are displayed in terms of number, surface and volume, and should be read by pairs, comparing the STAC observations to the control run (volcano-off) on the one hand, and the simulations including the volcano eruption (volcano-on) on the other hand.

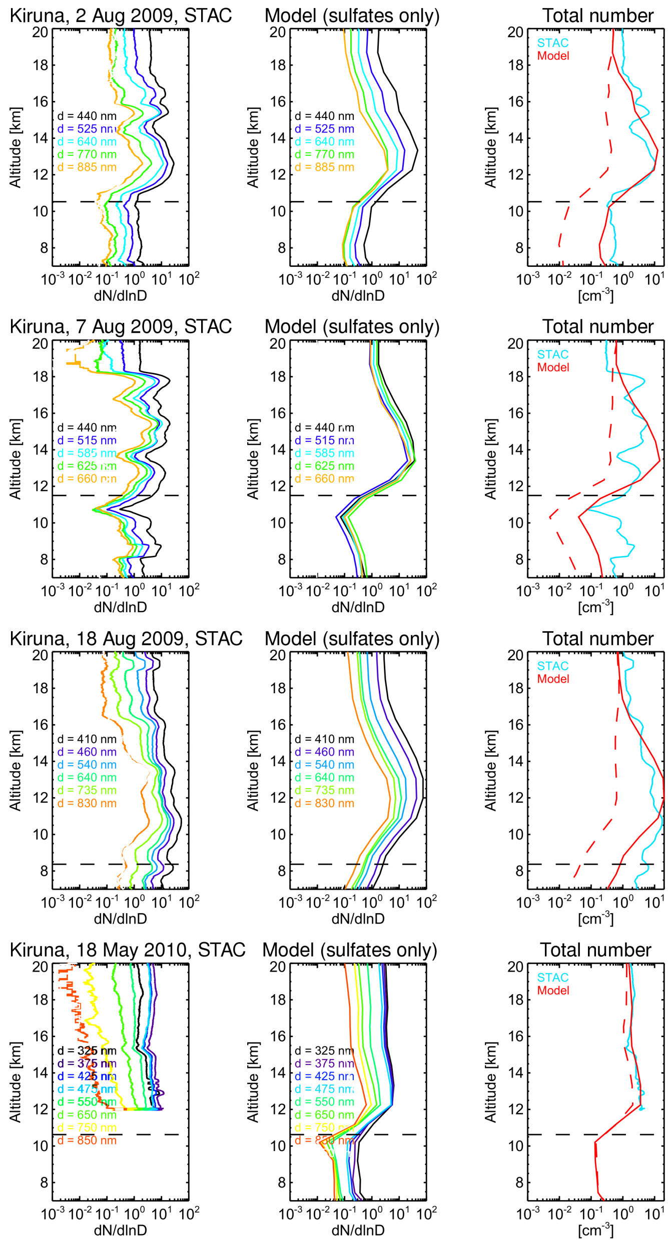

Figure 5. Comparison between STAC in situ measurements and CESM1(WACCM) simulations, over Kiruna, for 2, 7 and 18 August 2009 (plume detection) and 18 May 2010 (expected background conditions), and for altitudes ranging from 7 km to 20 km. Left column: particle counts operated by STAC, separated in size bins between 0.325 and 0.885 µm diameter. Second column: simulated equivalent through the use of the CESM1(WACCM) model. Third column: Comparison of the total particle counts for STAC and the model, over the STAC size-range. The red dashed line shows results from the simulation without volcanic aerosols. Error on the STAC total counts can be evaluated to be ±6%. The model tropopause altitude computed is represented by the horizontal black dashed line.

The figure highlights that the control run underestimates the particle number (area or volume) size distribution curves by orders-of-magnitude compared to the STAC observations. A much better agreement is found when the volcanic emission is included in the model simulations, showing a good ability of the model to reproduce volcanic aerosol plume in term of aerosol size distribution. For 18 May 2010, nearly one year after the eruption, the difference between the volcano-on simulation and control run is much less noticeable. This comparison reflects the ability of the model to simulate stratospheric aerosol size distributions in background (or near-background) conditions.

3.4 Comparison of the model SAOD to OSIRIS observations

Extinction data from OSIRIS have been used for model and observational assessment of stratospheric aerosol impacts following the Sarychev 2009 eruption (Haywood et al., 2010; Kravitz et al., 2011; O'Neill et al., 2012; Jégou et al., 2013). However, as mentioned in the Introduction, biases in the OSIRIS measurement following volcanic eruptions can affect the reported model-observation comparisons.

In Fromm et al. (2014), a detailed analysis of OSIRIS's limitations was carried out. These authors have shown that two main factors affect the derivation of SAOD by OSIRIS: (i) an upper detection limit on the value of extinctions, above which the measured values saturate; (ii) a latitude dependence in the minimal altitude above which extinctions are integrated to yield the SAOD.

As pointed out by Fromm et al. (2014), it is impossible to revert this process of data degradation; the best we can achieve to perform consistent model-to-observation comparisons is to degrade the extinctions derived from the model in order to derive SAOD "as OSIRIS would detect it". It must nonetheless be emphasised that such a comparison is not a complete evaluation of the model performance: any agreement found cannot fully validate aspects of the model output that are removed in the degradation process. Nevertheless, such a comparison of the degraded model to (biased) satellite observations is highly valuable: it enables an assessment model performance on a global-scale, which cannot be achieved using local-scale in-situ observations.

We use the following method: first, we allow the extinctions calculated in the model to saturate, with an upper threshold of 2.5×10^{-3} km^{-1} corresponding to the detection limit described in Fromm et al. (2014). Second, extinctions are integrated over truncated vertical columns of the atmosphere, introducing a lower altitude limit dependent on the latitude and the local tropopause height. Following Fromm et al. (2014), we define the minimal altitudes Z_{min} above which the extinctions are integrated as:

$$Z_{\text{min}}(\lambda, \phi, t) = Z_{\text{trop}}(\lambda, \phi, t) + \Delta(\phi) \quad (2)$$

where Z_{trop} is the local tropopause height, λ is longitude, ϕ latitude, t time, and Δ is a positive offset function, which was taken in our case as linearly varying with latitude from 0.5 km at the equator to 5.5 km at the poles. These were chosen as a trade-off between the histogram of values ~~(evaluated for 2012)~~ in Fromm et al. (2014), and actual minimum altitudes reached by OSIRIS ~~in 2009.~~ over the 2009–2010 period (see supplemementary material Fig. A2). For this series of calculations, ~~dynamical~~ thermal tropopause heights were diagnosed in the model. We verify the broad consistency of these altitude limits for OSIRIS

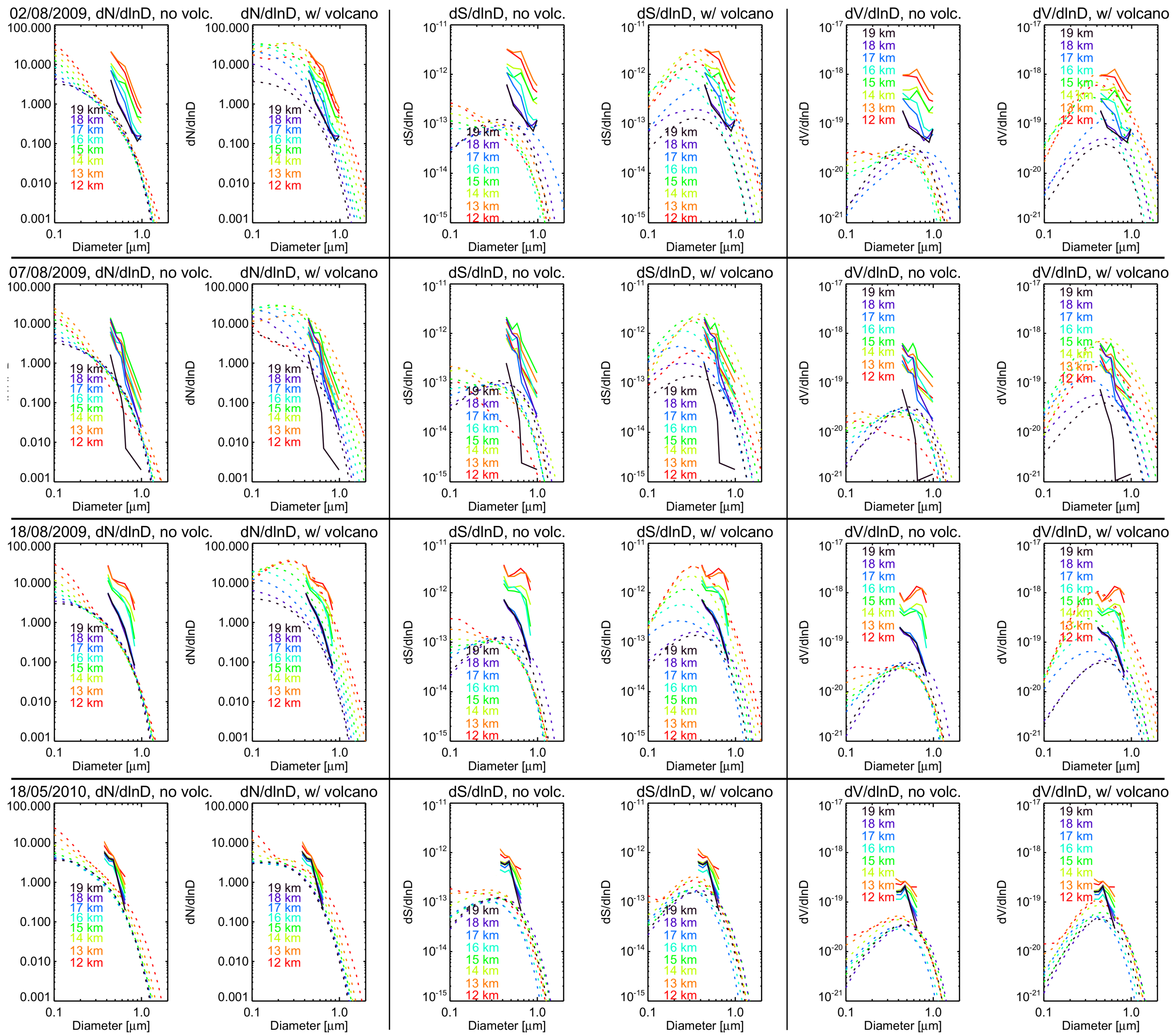

Figure 6. Particle size distributions in terms of number, area and volume, separated for different altitude layers, shown for the same four days of interest already presented in Fig. 5. Size distributions observed by STAC are shown as solid lines and simulated by CESM1(WACCM) as dashed lines. Graphs go by adjacent pair: comparison of STAC data to both the volcano-off and the volcano-on cases highlights the improved agreement between model and measurements for simulations when the volcano is active. The measurement error on STAC measurements can be evaluated to be ±6%.

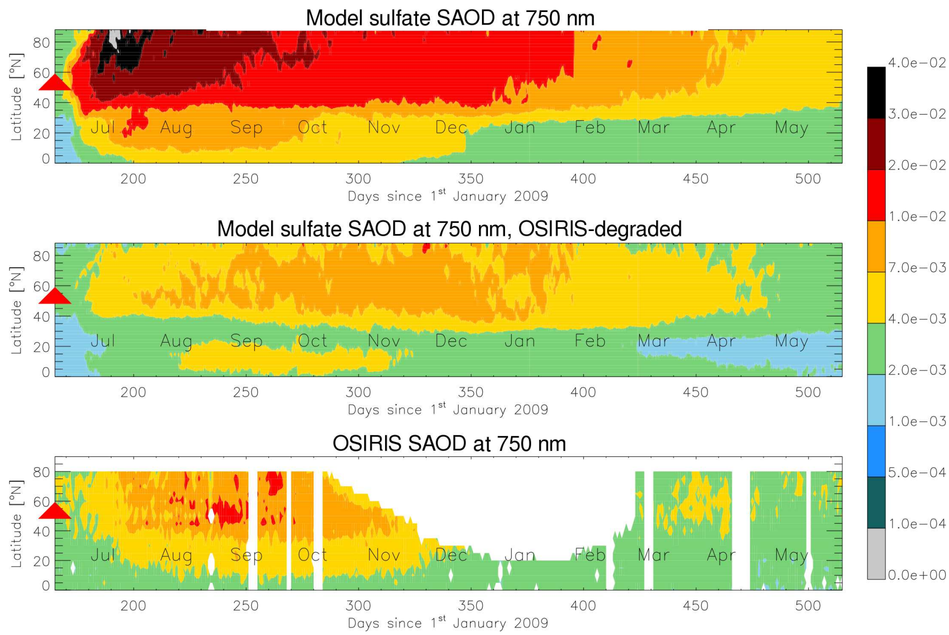

Figure 7. Comparison of modelled and observed stratospheric aerosol optical depth. Top panel: stratospheric sulfate aerosol optical depth at 750 nm as simulated by CESM1(WACCM). Middle panel: CESM1(WACCM)'s stratospheric sulfate AOD at 750 nm degraded to account for limitations in OSIRIS data (including saturation effect and minimum altitude). Bottom panel: actual OSIRIS SAOD retrieval obtained from data with measurement limitations. See text for details.

data during the 2009-2010 Sarychev post-eruption period in Fig.A2. Integrating the model-simulated saturated extinctions at 750 nm over the truncated altitude columns as defined above gives SAOD values that can be considered reasonably consistent with the measurements performed by OSIRIS.

Fig. 7 shows in the top panel the zonally averaged stratospheric sulfate AOD, through time, over the Northern hemisphere, as computed by CESM1(WACCM) in the volcano-on simulation (with co-injection of HCl). A degradation of the model data was then performed following the method described above. The resulting estimation of the sulfate SAOD "as would be detected by OSIRIS" is shown in the middle panel. The bottom panel shows the observed SAOD measured by OSIRIS. Over the winter months there is a lack of observational data from mid-October 2009 until the beginning of 2010, particularly at high latitudes that coincides with the polar night. A precise comparison for these months is therefore ~~difficult~~not possible. CESM1(WACCM) suggests that the sulfate SAOD remains at a fairly constant level over the Northern hemisphere over the October–December 2009 period, then decreases quite quickly from February 2010 to April 2010.

The degraded model SAOD shows reasonable agreement to the SAOD observed by OSIRIS, whilst the non-degraded model simulates much higher SAOD. This demonstrates that OSIRIS's limitations are crucial to the interpretation of its data. In Fig. 7, the observed (OSIRIS) SAOD shows, however, a slightly stronger maximal magnitude than the degraded SAOD from the model. A possible explanation may be that CESM1(WACCM) yields extinctions for sulfuric acid particulates only, whereas

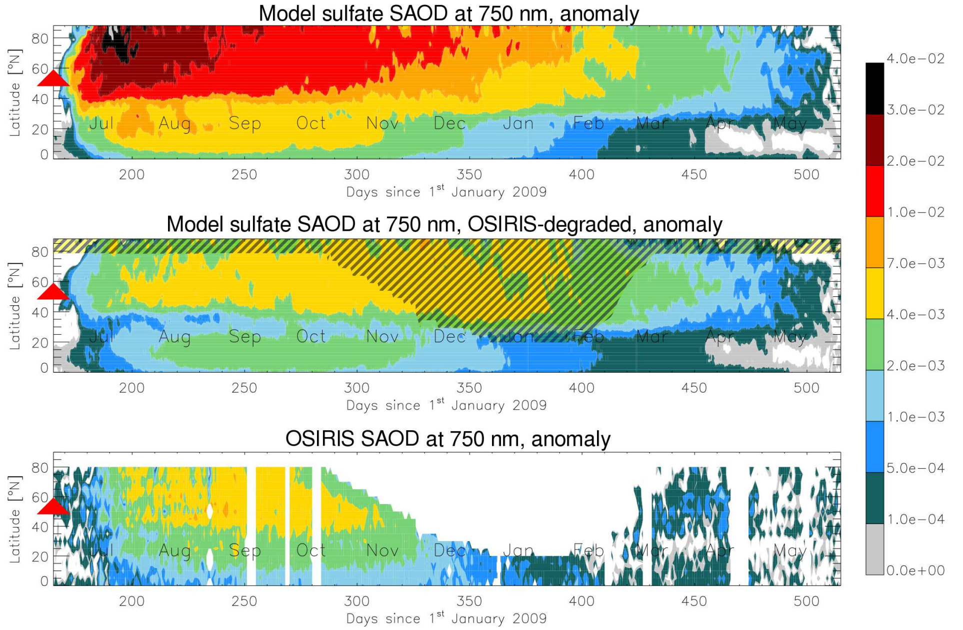

Figure 8. Comparison of modelled and observed anomalies in stratospheric aerosol optical depth. The absolute SAOD data from Fig. 7 have been converted to anomalies by subtracting modelled or observed SAOD one week before the eruption. Top panel: stratospheric sulfate aerosol optical depth anomaly at 750 nm as simulated by CESM1(WACCM). Middle panel: CESM1(WACCM)'s stratospheric sulfate SAOD anomaly at 750 nm degraded to account for limitations in OSIRIS data (including saturation effect and minimum altitude). Bottom panel: actual anomaly in OSIRIS SAOD retrieval obtained from data with measurement limitations. The shaded area denotes the polar night, where OSIRIS's measurements are missing. See text for details.

OSIRIS's observations account for a more comprehensive SAOD that can include non-sulfate compounds in the lower stratosphere.

To place a greater emphasis on sulfuric acid particulates due to the volcanic eruption, we convert all three datasets to anomalies. These anomalies were calculated by subtracting background conditions to the SAOD's, for which averages calculated on the first week of June 2009 were used as an approximate reference. Fig. 8 presents the same layout as Fig. 7, but now displays the SAOD anomalies over the same period (1 June 2009 until 31 May 2010). Again, a good accordance is found between the degraded model compared to OSIRIS (with the non-degraded model showing higher SAOD’’s). The agreement in SAOD anomalies in Fig. 8 is better than for the absolute SAOD's in Fig. 7. This indicates that differences in the background aerosol content prior to the eruption may explain some of the model-measurement discrepancy in term of SAOD maximum amplitude as highlighted in Fig. 7.

Integrating anomaly data presented in Fig. 8 yields the Northern hemisphere SAOD anomaly calculated at 750 nm over the year following the eruption, shown in Fig. 9. The dashed red line is SAOD simulated by the model. The plain red line is the same data after the OSIRIS bias degradation, and the blue line is SAOD from OSIRIS observations. Note that missing data in

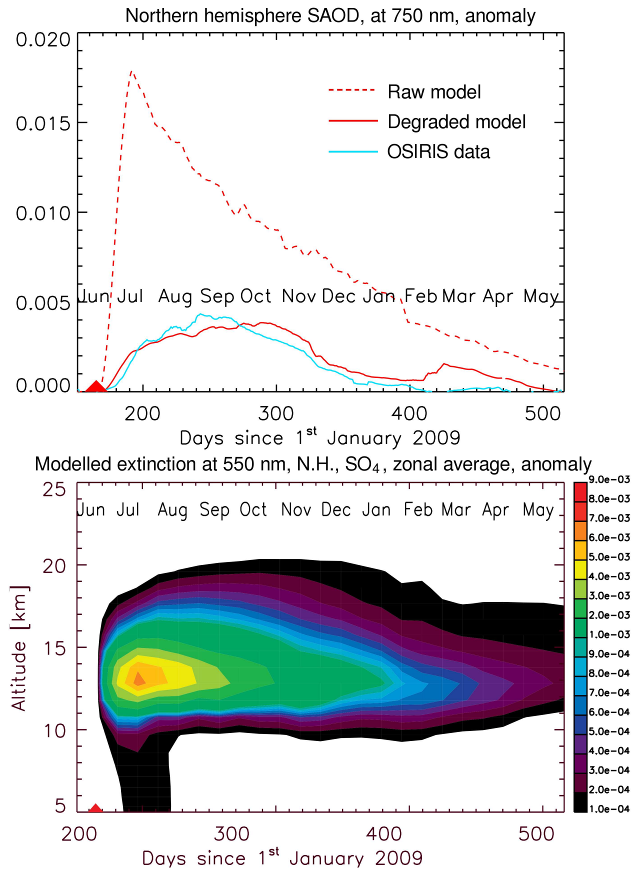

Figure 9. (Upper panel) Northern hemisphere SAOD anomalies at $750\,\mathrm{nm}$ calculated by integrating the model-simulated extinction (dashed red line), then degraded (full red line), and comparison with OSIRIS's actual data (blue line). The Sarychev eruption is symbolised by the red triangle. (Lower panel) Modelled temporal evolution of the sulfuric acid aerosol extinction coefficient at $550\,\mathrm{nm}$, zonally averaged for the Northern hemisphere, displayed in anomaly (volcano-on minus volcano-off).

OSIRIS's measurements during winter was taken into account in the integration of the degraded model data, as shown by the shaded area in Fig. 8. Fig. 8 along with Fig. 9 point out very clearly that taking into account OSIRIS's limitations gives a very good match between simulated and measured AOD values. The bottom panel in Fig. 9 shows the modelled temporal evolution in $550\,\mathrm{nm}$ extinction coefficients, zonally averaged for the Northern hemisphere, again highlighting maximum aerosol content around mid-July 2009.

Comparing the direct output of the model to OSIRIS in both Fig. 8 and Fig. 9 highlights a very much stronger and faster formation of sulfuric acid aerosols in the model than can be detected by the OSIRIS instrument, which experiences strongest measurement-biases shortly after the eruption due to the saturation effect. Further quantification is given below including e-folding times.

Reference	(Haywood et al., 2010)		(Kravitz et al., 2011)		Present study with CESM1(WACCM)		
Latitude band	HadGEM2	OSIRIS	ModelE	OSIRIS	Raw model	Degraded	OSIRIS
60°N to 80°N	60 days	66 days	57 days	81 days	99 days	45 days	41 days
40°N to 60°N	74 days	75 days	57 days	147 days	105 days	45 days	38 days
20°N to 40°N					120 days	49 days	31 days
0° to 20°N			60 days	408 days	65 days	41 days	41 days
Northern hemisphere	71 days	81 days			169 days	51 days	52 days

Table 3. SAOD e-folding times calculated for the model simulations and for OSIRIS's data reported in previous publications (Haywood et al., 2010; Kravitz et al., 2011) and in the present study. Different latitude bands are considered, and the decay times are all calculated considering SAOD values.

Analysing the temporal evolution of the (non-degraded) model SAOD identifies a peak in the Northern hemisphere 750 nm SAOD of ≈ 0.018 on 12 July 2009 (Fig. 9), followed by a long decay with an e-folding time of ≈ 169 days. Conversely, OSIRIS shows a much fainter and later peak (≈ 0.004 on 1 September 2009), with a quicker decay (e-folding time of 52 days). The degraded model SAOD yields an e-folding decay time (51 days) that is very comparable to that from OSIRIS; the peak value is also similar in amplitude to OSIRIS's and is reached on 17 October. This is slightly later than in the OSIRIS data, although the plateau in SAOD during that period can account for this delay.

Table 3 summarises the SAOD e-folding times calculated in this study, along with values from previous studies (Haywood et al., 2010; Kravitz et al., 2011), and calculated using OSIRIS data. Different bands of latitude are explored. One can note that the e-folding times vary quite significantly between authors, including those computed from OSIRIS's data (it is likely that different versions of the OSIRIS data—v.5.05 up to v.5.07 for the present study—were used). The main point to be highlighted here is the fair consistency obtained between e-folding times computed on the CESM1(WACCM)'s degraded data and on OSIRIS retrievals in our study, as evident from the last two columns of Table 3. We find that both the saturation limit and the fact that extinction profiles may terminate well above the tropopause are significant sources of measurement bias that need to be taken into account in comparison of OSIRIS data to model studies.

3.5 Post-eruption effective radius simulated using a sectional aerosol scheme

Discrepancies in the magnitude and e-folding times between model and OSIRIS SAOD’s have been mentioned elsewhere, notably in Haywood et al. (2010); Kravitz et al. (2011); O'Neill et al. (2012)previously mentioned, e.g. Haywood et al. (2010); Kravitz et a and are summarised here in Table 3. This led to a consequent questioning of the models' reliability to simulate sulfuric acid particle formation accurately in terms of timing, thought to be caused by the absence of nucleation of new particles in the model (Haywood et al., 2010; Jégou et al., 2013). Conversely, our study using the CESM1(WACCM) model, whose aerosol microphysics includes nucleation, finds very good agreement with OSIRIS retrievals of SAOD in terms of magnitude and temporally when the model SAOD is degraded to account for both saturation and minimum altitude limitations on the SAOD derived from OSIRIS measurements. The maximum in our (non-degraded) model SAOD is significantly higher (by a factor ≈ 4.5) than estimated by both OSIRIS and earlier modelling studies of the 2009 Sarychev Peak eruption (Haywood et al., 2010). A key unconstrained parameter in these earlier studies was the stratospheric particle size distribution that exerts a strong influence on SAOD. It was set to yield an effective radius of around $r_{\mathrm{eff}} = 0.13$–$0.15\ \mu\mathrm{m}$ in Haywood et al. (2010), with the model results from Kravitz et al. (2011) also adjusted to represent this size. Previous studies suggested higher r_{eff} for large magnitude eruptions that injected SO_2 higher into the stratosphere (yielding longer-lived sulfate clouds): Russell et al. (1993) derived r_{eff} of $0.22 \pm 0.06\ \mu\mathrm{m}$ around one month after the Mt. Pinatubo 1991 eruption, whilst Stothers (1997, 2001) suggest post-eruption r_{eff} grew from around 0.2–0.3 to 0.4–0.5 $\mu\mathrm{m}$ over the time-scale of one year. Conversely, a lower r_{eff} was thought to be reasonable for the moderate-magnitude 2009 Sarychev Peak eruption that injected to the lower stratosphere (yielding relatively fresh and shorter lived sulfate cloud), and appeared consistent with ground-based remote sensing at Mauna Loa (Hawaii, U.S.A.) (Barnes and Hofmann, 2001; Haywood et al., 2010).

Here, the sectional aerosol representation with full aerosol microphysics in CESM1(WACCM) enables to freely simulate the post-eruption evolution in particle size, without any a priori assumptions. Sulfuric acid is first produced by the oxidation of volcanic SO_2, which leads to formation of new sulfuric acid particles by nucleation. Processes such as particle coagulation and condensation of sulfuric acid onto the existing particles causes particle growth. Particles are removed from the stratosphere by sedimentation and tropopause folding (Hamill et al., 1997). The balance between these processes determines the overall size distribution and its effective radius. Fig. 10 shows the zonally averaged effective radius simulated by the model for three latitude bands (20°N to 40°N, 40°N to 60°N and 60°N to 80°N). Particle growth occurs in regions with elevated sulfate following the volcanic eruption (Fig. A3, supplementary Materialmaterial). Particle size grows to reach a maximum in zonal mean r_{eff} of up to $0.2\ \mu\mathrm{m}$ in the lower stratosphere. The greatest enhancement in r_{eff} occurs at high latitudes as expected given the poleward atmospheric transport in the stratosphere. At mid-latitudes, a temporary decrease in r_{eff} can also be seen immediately following the eruption: this is due to new particle formation (nucleation) of particles of a few nm-size. The latitudinal trend in r_{eff} simulated by our model is broadly consistent with the trend reported from ground-based remote sensing at Eureka (Nunavut, Canada) that found $r_{\mathrm{eff}} = 0.29\ \mu\mathrm{m}$ (O'Neill et al., 2012), and with ACE measurements, which report $r_{\mathrm{eff}} = 0.1 - 0.3\ \mu\mathrm{m}$ (Doeringer et al., 2012). Modelled absolute values of r_{eff} are also globally consistent with balloon-borne observations in August 2009 (Jégou et al., 2013). Aerosol size or r_{eff} exerts a strong influence on SAOD (e.g. Haywood et al. (2010)). A priori assumptions in stratospheric particle size are a thus thus a major source of uncertainty in model studies that do not freely simulate the aerosol size-evolution, and that will tend to cause an underestimation of SAOD in cases where the assumed r_{eff} is lower than reality.

3.6 Effects of SO_2 and HCl co-injection on stratospheric chemistry

Most studies investigating impacts from modern-day eruptions ' the impacts of modern day eruptions on stratospheric chemistry have focused on the role of sulfuric acid particles in reducing NO_x levels and activating pre-existing chlorine and bromine (ClO_x, BrO_x) in the stratosphere (Fahey et al., 1993; Solomon, 1999). One must note that halogens from the 1991 Mt. Pinatubo eruption were efficiently washed out and therefore did not reach the stratosphere (Mankin et al., 1992; Tabazadeh and Turco, 1993), though the washout for the Sarychev case was not necessarily as efficient (von Glasow et al., 2009). Observational ev-

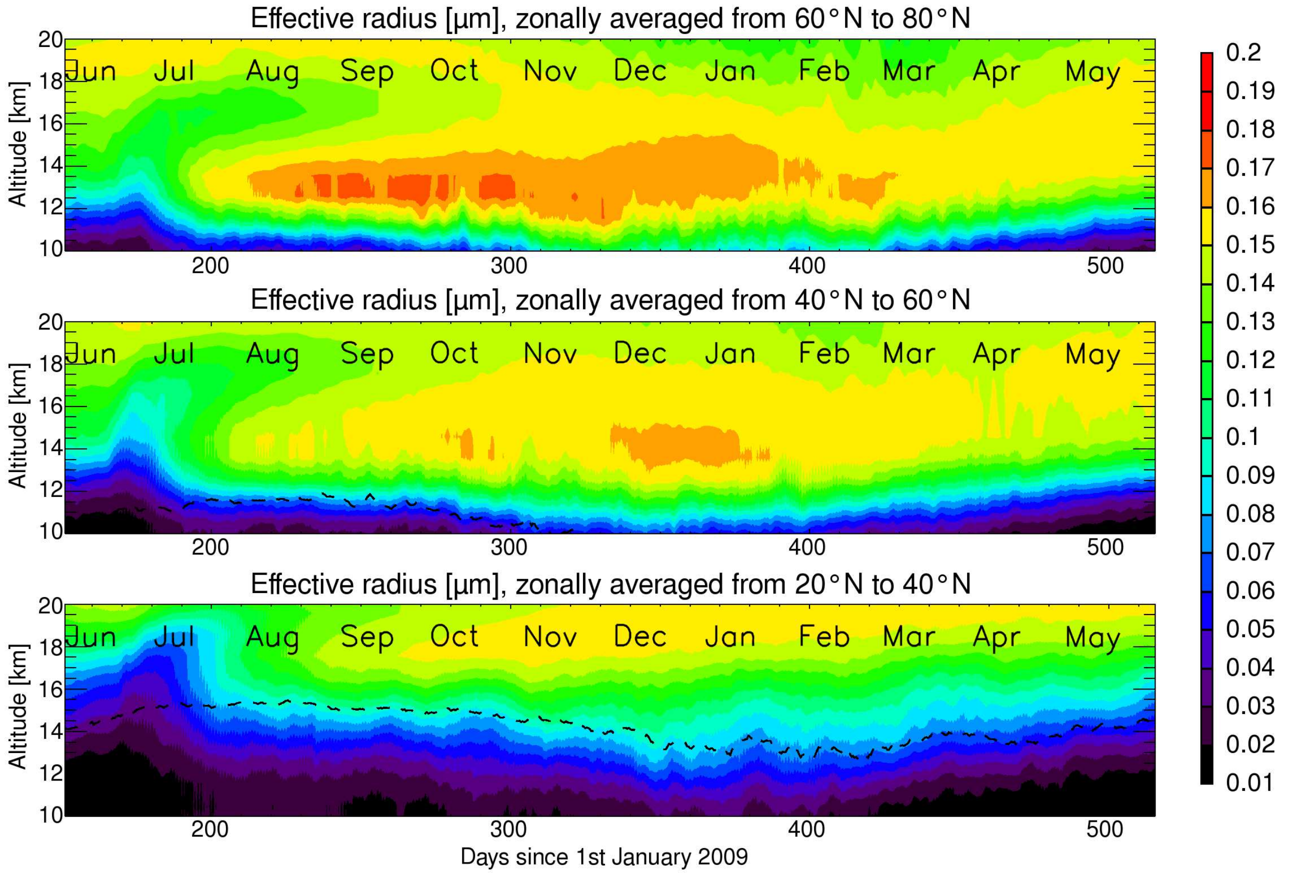

Figure 10. Zonally averaged effective radius simulated by CESM1(WACCM) model, in μm , as a function of altitude for three latitude bands (20°N to 40°N, 40°N to 60°N and 60°N to 80°N). The model tropopause is shown as a dashed line.

idence of stratospheric NO_2 depletion following moderate-magnitude volcanic eruptions is provided by Adams et al. (2017) based on satellite remote sensing, and Berthet et al. (2017) by balloon-borne observations following the Sarychev Peak eruption. Our study builds on these recent works in two aspects: first by using the CESM1(WACCM) model with sectional aerosol representation we freely-simulate the aerosol surface area (SAD) (a function of particle number and size) that is a key control on stratospheric chemistry impacts. Second, we investigate the stratospheric chemistry influence of volcanic HCl that observations show was co-injected alongside SO_2 (Carn et al., 2016). The simulated anomalies in ozone and NO_2 for the latitudinal bands 40°N to 60°N and 60°N to 80°N are shown for simulations with SO_2 injection only, and for HCl co-injection with SO_2 in Fig. 11. In summer, greater depletions of up to -60% for NO_2 are found at higher latitudes. This is primarily due to the ~~latitudinal extent of the volcanic cloud with~~ higher aerosol loadings in these regions, ~~keeping however in mind that reduction saturates at a certain level of SAD (Fahey et al., 1993), and to favored~~ and to favorable solar illumination conditions for which the catalytic ozone loss cycles (through OH radical production) are enhanced (Berthet et al., 2017). ~~As the season~~

progresses more is converted to the nitrogen reservoir at nighttime with decreasing solar illumination conditions (especially at high latitudes) and the subsequent conversion to the more stable reservoir by enhanced hydrolysis of on volcanic aerosol sequesters more at higher latitudes. As the polar vortex builds up, decreasing temperatures in the high latitude stratosphere favor chlorine activation by temperature-dependent heterogeneous processes (mainly involving) leading to denoxification (i. e. loss of by enhanced reaction of with and by conversion of the nitrogen reservoir to more stable) and ozone depletion (Solomon, 1999). A more detailed description of the involved chemical processes is provided in Berthet et al. (2017).

Our results are broadly consistent with the observations of Adams et al. (2017) who reported that stratospheric NO_2 abundances were reduced by up to ≈45–55% over 40–80°N as consequence of the Sarychev Eruption. Berthet et al. (2017) showed maximum NO_2 depletion of ≈50% in the summertime lower stratosphere above Kiruna (Sweden) following the Sarychev eruption, based on REPROBUS model simulations with SAD prescribed from observations and without HCl injection. They predicted ozone depletion reached up to 4% in the summertime/early fall lowermost stratosphere. Our CESM1(WACCM) simulation with a sectional aerosol scheme and injection of volcanic SO_2 (only) finds similar high latitude maximum NO_2 reduction (≈ 50%) and maximum ozone depletion (5%). Interestingly, our simulations suggest somewhat greater maximum depletions in the simulation with co-injected volcanic HCl (7% and 60%, for ozone and NO_2 respectively) compared to the simulation with SO_2 injection only (5% and 50%, for ozone and NO_2 respectively). As a result of the enhanced stratospheric HCl budget throughout the season more ozone-depleting chlorine radicals are expected to be formed due to reaction R1 even at mid-latitude conditions, though the impact on ozone appears limited. The impact on summer and fall NO_2 is negligible. In the polar winter, cold temperatures lead to further chlorine activation through heterogeneous processes enhancing some NO_x and ozone reduction. This study highlights the potential for volcanic HCl to supplement and enhance SO_2-sulfate impacts on stratospheric chemistry, for eruptions where there is a significant HCl injection to high altitudes. The influence of Sarychev Peak eruption on stratospheric chemistry is nevertheless relatively modest, due to the moderate eruption size, in terms of both the SO_2 and HCl injected amounts.

4 Conclusions

We have presented a series of simulations carried out with the CESM1(WACCM) model, for the study of stratospheric chemical impacts from the moderate-magnitude 2009 Sarychev eruption. Associated with the CARMA module, the model explicitly simulates the aerosol size evolution using a sectional aerosol scheme (across 30 size-bins), and includes detailed aerosol microphysics. To simulate the eruption, we assumed a 0.9 Tg injection of sulfur dioxide between 11 km and 15 km altitude over the one day of eruption (15 June 2009). We also investigated the impacts of co-injected volcanic HCl.

Through comparison of the model results with satellite (IASI) retrievals of SO_2 and in situ measurements of stratospheric aerosols, we were able to assess the model performance, finding good agreement in terms of plume dispersion (Fig. 1) and particle formation rates (Fig. 2, Fig. 3), particle number concentrations as well as particle size distributions before and following the eruption (Fig. 4, Fig. 5, Fig. 6). In particular, very good agreement was found in terms of particle number concentrations and particle size distributions obtained from balloon-borne observations over Kiruna (Northern Sweden) in July-August 2009

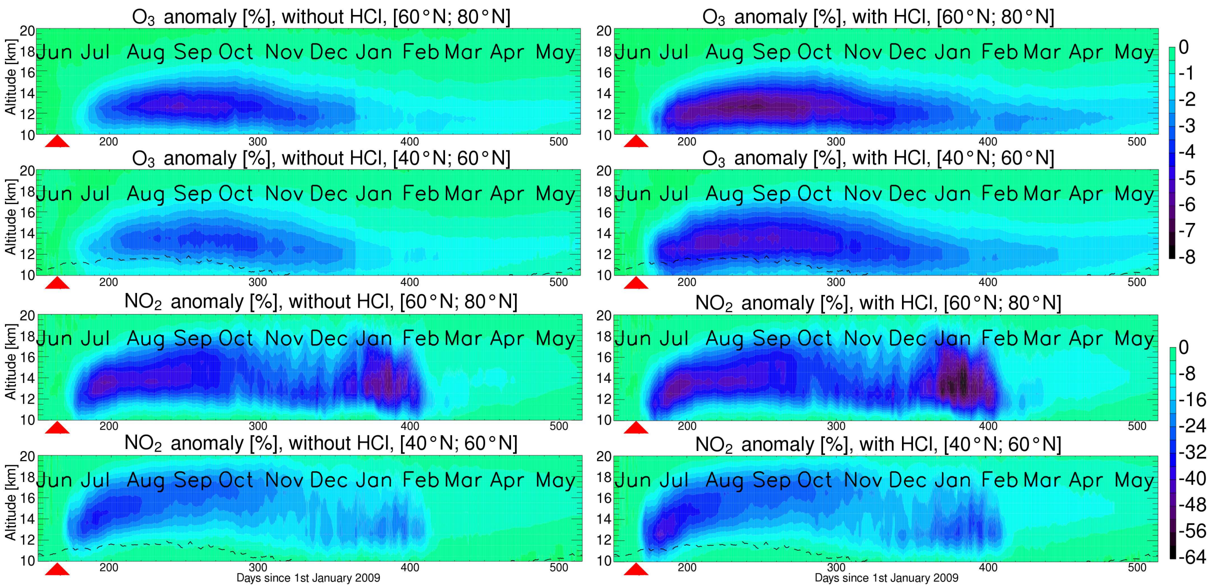

Figure 11. Zonally averaged depletions in stratospheric ozone and NO_2 at mid- (40°N to 60°N) and high-latitudes (60°N to 80°N) following the Sarychev eruption. Ozone is shown in the upper set of plots, NO_2 in the lower sets; within each pair, the latitudes are shown as: 60°N to 80°N (upper plot of pair) and 40°N to 60°N (lower plot of pair). Simulations by the CESM1(WACCM) model are expressed as percentage anomalies (with respect to the volcano-off control run) and are calculated for the simulation with SO_2 injection only (left), and simulation with co-injection of HCl (right). Impacts on stratospheric chemistry are greater at higher latitudes and are enhanced by co-injection of volcanic HCl.

(Fig. 5, Fig. 6) and Laramie (Wyoming, USA) in November 2009 (Fig. 4) confirming the strong impact of the volcanic eruption on the stratospheric aerosol particle load. This suggests that particle formation is represented well in the sectional aerosol scheme (CARMA) in CESM1(WACCM). The simulations suggest that the effective radius (r_{eff}) becomes enhanced following the eruption to reach up to $0.2\ \mu m$ in the zonal average. This is larger than the fixed aerosol size assumed in previous model studies with limited aerosol microphysics, e.g. $r_{eff} = 0.13–0.15\ \mu m$ (Haywood et al., 2010). ~~This~~ The lack of more resolved data might be a source of uncertainty on the injection altitudes. However, this overall quantitative agreement ~~lends support to the~~ reflects the model performance in SO_2 oxidation, atmospheric dispersion and aerosol processing. It indicates a suitable choice of eruption source parameters as used in previous studies ~~:~~ e.g. Haywood et al. (2010) (an injection altitude ranging from ~~to~~ 11 km to 15 km for SO_2, ~~as was already suggested and used in Haywood et al. (2010), appears to be realistic. Likewise, the assumption of~~ a vertical even spread of the total mass of gases injected, and a sole injection of the total gas mass on 15 June 2009, neglecting other minor injections on other days~~,~~). These eruption source parameters did provide good results. ~~We also~~ They might need to be refined for model studies at higher temporal or spatial resolution, see Wu et al. (2017), Günther et al. (2017). We point out that an injected mass of 0.9 Tg SO_2 (Clarisse et al., 2012; Realmuto and Berk, 2016) instead of 1.2 Tg of previous studies e.g. Haywood et al. (2010) is a fair hypothesis, and enables the model to closely reproduce the observed SO_2 burden according to the IASI retrievals of Clarisse et al. (2012).

In addition, we investigated the co-injection of volcanic HCl to the stratosphere. We based our simulations on reported stratospheric HCl/SO_2 mass ratio of 0.03 for Sarychev Peak eruption, according to analysis of satellite data by Carn et al. (2016). The altitude and timing of the HCl injection in the model were assumed to be identical to the SO_2 injection. Our study suggests that the presence of HCl leads to a delay in the oxidation of SO_2 to form sulfuric acid particles of about two days, with a 5–10% increase in the modelled e-folding times for SO_2. We also find a better temporal accordance in SO_2 burden derived from satellite (IASI) data and our simulations when taking HCl into account. The additional surface area provided by volcanic particles catalyses reactions that can perturb stratospheric chemistry, including activation of stratospheric halogens, and can lead to strong reduction of NO_2 and modest depletion of ozone as highlighted by Berthet et al. (2017) for Sarychev Peak. Our simulations show that the co-injected volcanic HCl also affects the post-eruption stratospheric chemistry of ozone and NO_x, depleting these species more severely than in simulations that account for SO_2 injections only. Our results highlight that volcanic HCl emissions should be taken into account when simulating sulfur chemistry and stratospheric chemistry impacts from volcanic eruptions during which HCl is co-injected.

The second major point highlighted by this paper is the treatment of limitations in SAOD derived from OSIRIS measurements: both a saturation effect and a varying minimum altitude in available OSIRIS data (i.e., extinction profiles may terminate well above the tropopause in particular at high latitudes) were identified by Fromm et al. (2014). We used a two-step model degradation process to reproduce these biases in the modelled data, and found as a result very good agreement with the actual OSIRIS measurements following the volcanic eruption, reproducing both the magnitude and temporal evolution of the SAOD following the 2009 Sarychev eruption (Fig. 7, Fig. 8, Fig. 9, Table 3). Recent studies (Haywood et al., 2010; Kravitz et al., 2011; O'Neill et al., 2012) quantifying volcanic impacts have tended to (only) incriminate their models' particle formation schemes because the comparisons with OSIRIS's satellite retrievals were poor specifically regarding the timing of the SAOD maximum. As a matter of fact, caveats on OSIRIS's measurement, as outlined in Fromm et al. (2014), are the key point to any model-observation comparison. We show that there is a considerably improved match between simulated and observed SAODs when these are taken into account. Once again, we stress that this accordance is only obtained by degrading the model output to account for OSIRIS's caveats; the fact that Fig. 8 and Fig. 9 show similar anomaly values cannot be sufficient to thoroughly validate the modelled SAOD's through time. Rather, they provide supportive evidence to our study of stratospheric aerosol evolution following the Sarychev Peak 2009 eruption, using a model with detailed aerosol microphysics and sectional aerosol representation. The non-degraded output from our model shows substantially higher SAOD (maximum of 0.018 at 750 nm) than observed by OSIRIS (0.004), or as reported by previous model studies (with fixed aerosol size, and limited microphysics). Our study therefore highlights that previous modelling studies (involving assumptions on particle size) that reported agreement to (biased) post-eruption estimates of SAOD derived from OSIRIS likely underestimated the climate impact of the 2009 Sarychev Peak eruption

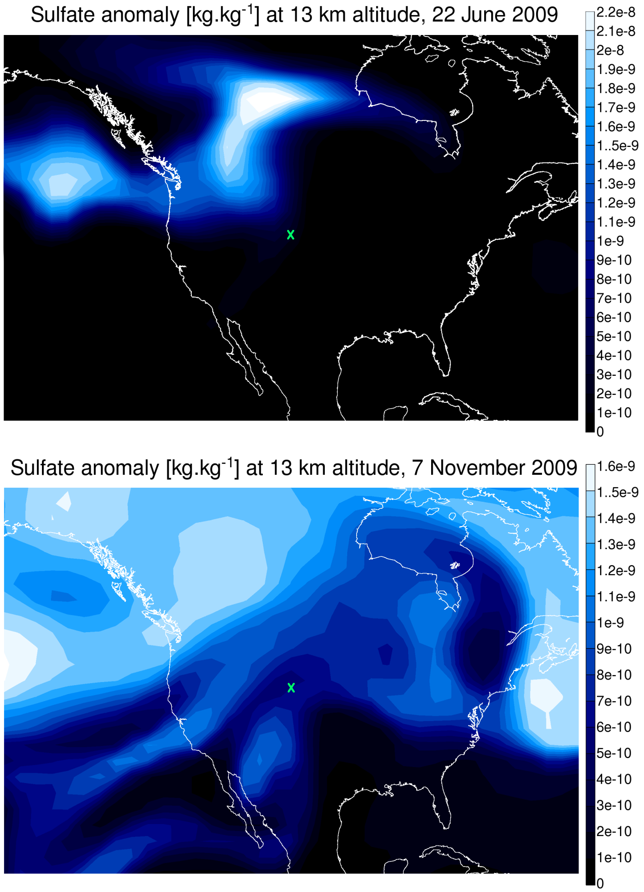

Figure A1. Geographic map over the U.S.A. displaying the simulated sulfate aerosol at 13 km altitude on 22 June 2009 (upper panel) and 7 November 2009 (lower panel), as computed by CESM1(WACCM). Note order of magnitude difference in colour scale between the two plots. Laramie is indicated by the green cross: it is located on the very edge of a modelled aerosol plume structure on 22 June 2009, but below a more wide-spread (and dilute) plume on 7 November 2009.

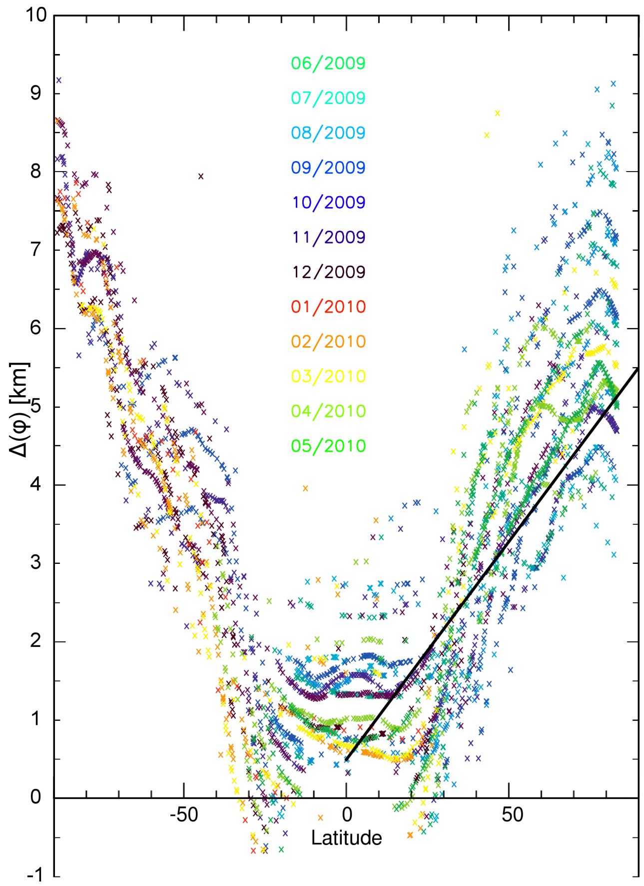

Figure A2. Minimum altitude of OSIRIS extinction data calculated relative to the model tropopause as a function of latitude. OSIRIS data are shown for the first of every month from June 2009 to May ~~2010.~~ 2010, with different colours depending on the month considered. There is some missing data during the winter in each hemisphere, particularly at high latitudes. The solid line shows the latitude dependence of the minimum altitude threshold assumed in the model degradation in this study.

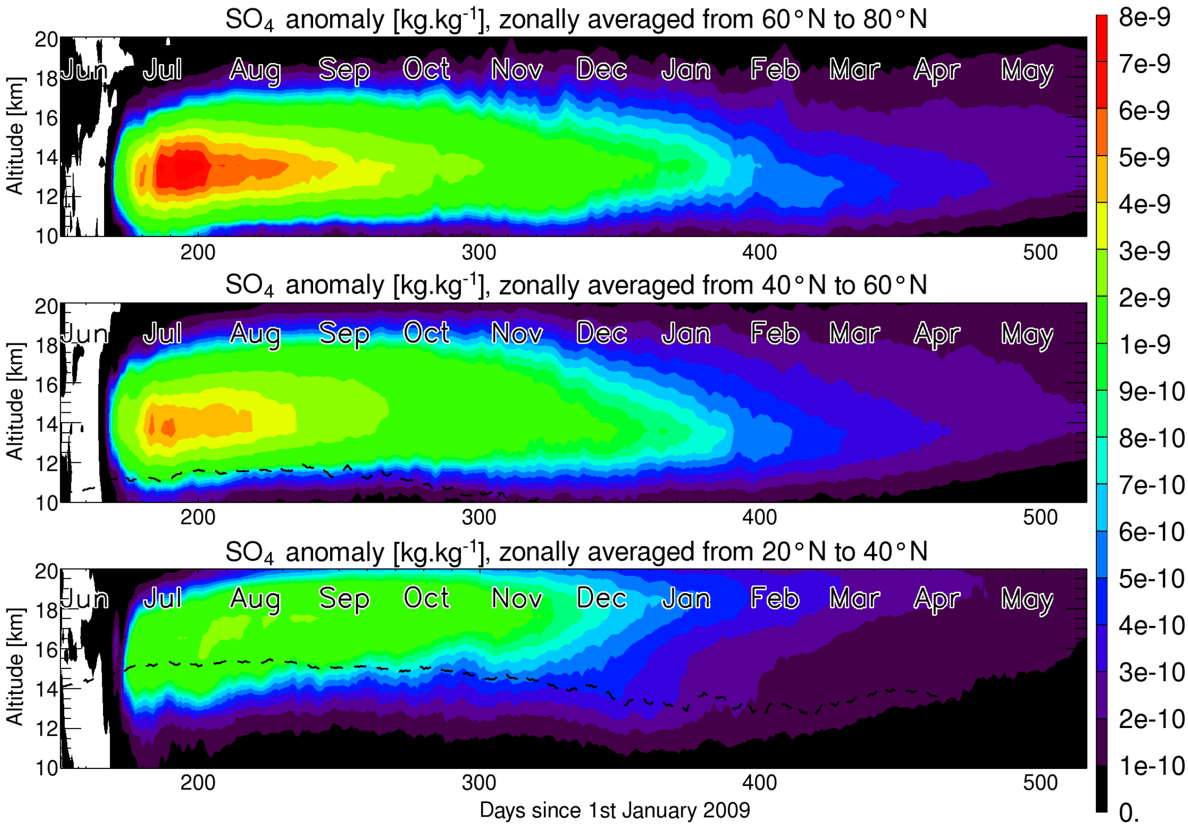

Figure A3. Zonally averaged sulfate anomaly, in kg.kg^{-1} , as simulated by CESM1(WACCM) model as a function of altitude for three latitude bands (20°N to 40°N, 40°N to 60°N and 60°N to 80°N). The model tropopause is shown as a dashed line.

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