Final author response to the Referees' comments on "Sensitivity of the radiative forcing by stratospheric sulfur geoengineering to the amount and strategy of the SO_2 injection studied with the LMDZ-S3A model"

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We thank the four Referees for their thorough evaluation of the manuscript. Their comments are repeated below in blue and our response follows in black.

Response to comments by Referee 1

This manuscript reports on studies of geoengineering via stratospheric SO2 injection performed with the GCM LMDZ coupled to a sectional aerosol model S3A (the coupled model is called LMDZ-S3A). The model has some limitations (fixed SST, no ozone feedbacks, fixed chemical time scale for SO2 conversion, fixed aerosol composition for radiative calculations) but nevertheless does include important feedbacks (aerosol feedback on radiation, interactive tropopause height, self-generated QBO) for stratospheric aerosol injection (SAI). The paper is generally well-written and the subject matter appropriate to ACP. This paper repeats some experiments of a study performed by Niemeier and Timmreck (2015) but with a different model, and interestingly it reaches somewhat different conclusions. Given the uncertainties in global modeling of something like stratospheric aerosol injection, having model results from multiple independently-formulated models is very desirable. And when models diverge in their conclusions, it may lead us to widen the uncertainty bounds, but also to investigate mechanism, feedbacks, and model reliability.

I support publication of this paper after the following scientific issues are addressed.

We thank the referee for his/her appreciation and the accurate summary of our study.

Specific Comments:

I would like the paper to emphasize somewhat more that the results presented apply only to injection of SO2 – injection of H2SO4 (see Pierce et al. (2010)) or even injection of SO2 outside the tropics, may display different RF responses and scalings. In several places the paper uses "stratospheric sulfate aerosol" or "sulfate SAI" to refer to SO2 injection with statements that may not be as appropriate for injection of H2SO4 or preformed sulfate aerosol particles. The language just needs to be more precise (such as "tropical injection of SO2") to avoid overgeneralizing the conclusions.

We agree with the reviewer and have corrected the manuscript to be more precise on the injection techniques tested in the study.

The reference Boucher et al. (2017), referred to as a "companion paper" in the introduction, is incomplete. Perhaps this article is currently under review, as searches of GRL do not reveal such a paper. Given that this reference contains analysis of the importance of rapid adjustment of T, H2O and cloud fields to RF, it should have been included with the review materials.

We apologize to the reviewer if this has hindered his/her review in some way. We would have been happy to communicate the submitted paper to the reviewer via the ACP editorial office. In any case this "companion" paper is now published under doi:10.1002/2017GL074647.

It would be useful to show a comparison of radiative forcing generated, not only as the difference in a single simulation calculated with and without aerosols, but also as the SAI scenario minus the CONTROL scenario. This would allow the reader to know the magnitude of the impact of changes in the temperature and H2O on RF. The other part of the RF calculation which should be quantified is the portion of the calculated RF due to background sulfate, which is apparently not subtracted in the current methodology. Calculating RF of the CONTROL case with and without aerosols would provide that.

The background sulfate aerosol has a global and annual mean net instantaneous radiative forcing of –0.04 $\mathrm{W\,m^{-2}}$ (–0.05 $\mathrm{W\,m^{-2}}$ in the SW and +0.01 $\mathrm{W\,m^{-2}}$ in the LW). We added this information to the text, but do not subtract it from the presented RF values of SAI, since it may be considered as negligible.

Text describing Figure 12 is missing. Perhaps an accidental deletion of text on page 10 between "The particle size. . .appears to approach saturation levels below 0.5 um with increasing injection height." And "Only the aerosol mass on the lower end of the size range decreases further. . .". The latter part of this paragraph seems to refer to Figure 12.

Thank you for pointing this issue. We added a reference to Figure 12 to this sentence.

The "Conclusion" section should mention the lack of chemical feedbacks through ozone.

We added a sentence on chemical feedbacks and ozone to the conclusions.

Technical Corrections:

Page 8, line 23: "monotonously" should be "monotonically".

Done.

In Figures 7 and 10, the colors for LW and SW forcing are different shades of blue which are difficult to distinguish. Why not different colors rather than shades of the same color? Also, a legend on the plots describing the different colors and symbols would aid comprehension.

The choice of colours is always difficult and is often a matter of personal preferences. We chose these colours as they were already used by Niemeier and Timmreck (2015) to illustrate SW, LW and net radiative forcing.

Response to comments by Referee 2

This paper describes the use of the LMDZ-S3A model to study the efficacy and impacts of stratospheric sulfur geoengineering. This is valuable research, as stratospheric aerosols from many sources (e.g. volcanoes and hypothetical geoengineering schemes) can impact climate and chemistry, and there remains much uncertainty regarding stratospheric processes and aerosol

evolution. This is an excellent model for this work, as the model includes aerosol microphysical processes, sectional aerosol size representation, aerosol-radiative interactions in both the SW and LW, high vertical resolution, stratospheric processes including QBO, and precursor gas emissions. The authors apply this model to study the impacts of equatorial SO2 injections at various magnitudes (2-50 Tg S/yr) and altitudes (15-23km) as well as a seasonal and broad injections, on aerosol properties, radiative fluxes, stratospheric circulation, and surface pollution. The paper is well-written, clearly laid out, and provides valuable contributions to scientific understanding of stratospheric sulfur geoengineering. I have one general/moderate concern and numerous small/specific concerns with the manuscript.

We thank the reviewer for his/her positive comments.

General/moderate concern:

The complexity of aerosol-chemical-dynamic processes, combined with long aerosol lifetime in the stratosphere, results in high uncertainty with models. Indeed, the results of this work differs fairly significantly from some other work, which the authors cite (for example, the large differences in radiative efficacy at higher injection rates compared to Niemeier and Timmreck (2015), and the large differences in efficacy at higher injection altitudes compared to Niemeier et al. (2011); Niemeier and Timmreck (2015)). As such, I recommend a few things:

1) Add a paragraph to the introduction, as well as the conclusion, highlighting the significant uncertainties with stratospheric processes, especially hypothetical geoengineering schemes which have no observations to evaluate models against (yet)

We added the following sentence: "The stratospheric processes determining this are complex and uncertain and their implementation causes considerable disagreement between different models. In contrast to volcanic eruptions, models cannot be constrained by observations in the case of purely hypothetical geoengineering scenarios."

2) Add more discussion throughout the paper on possible sources of uncertainties and errors with the current model implementation.

A few discussion items:

a) Prescribed oxidants: How do you know that prescribed oxidants in your model do not significantly impact results? It would be nice to do a sensitivity study, perhaps varying the availability of oxidants, and calculating how it impacts results. How might its feedback with water vapor impact results? Why did you not quantify ozone destruction, which is a known possible consequence of stratospheric sulfur geoengineering? Discuss.

We performed a sensitivity study for the prescribed chemical lifetime of SO_2 under Pinatubo conditions in Kleinschmitt et al. (2017) (see Fig. 14 therein). A doubling of the lifetime on the day of the eruption, decreasing linearly to normal values within 1 month, had a relatively small impact on the resulting AOD by delaying and increasing the peak AOD slightly. The impact can be expected to be even smaller for scenarios of continuous SAI with moderate SO_2 injection rates, as they result in lower local SO_2 concentrations than a sudden major volcanic injection of SO_2.

The impact of SAI on stratospheric water vapour is discussed in Boucher et al. (2017). The greenhouse effect of the additional water vapour (1 to 3 ppmv in the STANDARD experiment) partly compensates the negative radiative forcing of the aerosol. As a feedback on stratospheric chemistry is not included in our simulations, we cannot quantify ozone destruction.

We added the following sentence to Sect. 2.1 discussing the issue of prescribed oxidants: "For continuous SAI schemes, where

the aerosol layer and its properties reach an equilibrium after several years, we would expect oxidants to reach equilibrium concentrations (somewhat) lower than the climatological values. Assuming a climatological chemical lifetime of SO_2 could therefore cause the model to overestimate SO_2 to H_2SO_4 conversion rates to a certain degree, favouring new particle formation over condensational growth."

b) Coagulation processes: There are numerous coagulation processes that might be significant for geoengineered stratospheric aerosols – for example, you mention van der Waals forces. As you note, previous studies have found van der Waals forces to impact ambient stratospheric aerosol, and volcanic eruptions. The impacts of van der Waals forces on geoengineered aerosols seems uncertain, and possibly important. I suggest conducting an experiment with van der Waals forces compared to the standard experiment to see if it impacts results; at a minimum, further discussion.

In Kleinschmitt et al. (2017) we also performed a sensitivity study for the impact of van der Waals forces under Pinatubo conditions (see Figs. 12 and 13 therein). We found that including them slightly improved the agreement of the modelled particle number and size with observations, but deteriorated the agreement in AOD. Given these mixed results and the uncertainty of the van der Waals coagulation term, we chose not to include this effect in the SAI experiments.

However, for the avoidance of doubt we performed additional experiments where the van der Waals (vdW) effect on coagulation is included. The results show that vdW enhancement may further increase particle size and considerably reduce the radiative forcing for larger injection rates. This is now discussed in the manuscript in the new Section 3.5 ("Sensitivity to van der Waals coagulation enhancement factor").

c) Any other limitations in your model worth discussing further?

The use of fixed sea surface temperatures (SSTs) might be another limitation (see response to Referee 4). Therefore we added the following sentence to the conclusions: "The prescription of sea surface temperatures constitutes another limitation for correctly simulating the feedback of SAI on atmospheric dynamics."

Specific comments:

Abstract:

1) More clearly state that you calculate the limit of sulfate geoengineering efficacy to be 2 Wm-2, and that efficacy actually decreases at injections larger than 20 Tg S/yr – this is very interesting.

We modified the sentence in the following way: "As a result, the net instantaneous radiative forcing does not exceed the limit of –2 $\mathrm{W\,m^{-2}}$ for continuous equatorial SO_2 injections and it decreases (in absolute value) for injection rates larger than 20 $\mathrm{Tg\,S\,yr^{-1}}$."

2) Add a caveat that stratospheric processes are complex and uncertain and that multiple modeling studies disagree with one another, including your model.

We added the following sentences: "But it has to be noted that the complexity and uncertainty of stratospheric processes cause considerable disagreement between different modelling studies of stratospheric aerosol geoengineering. This may be addressed through detailed model intercomparison activities, as observations to constrain the simulations of stratospheric aerosol injection are not available and analogues (such as volcanic eruptions) are imperfect."

p1, line 20: suggest that you change the term "physico-chemistry" to something more understandable. Also, does this include

impacts of ozone? If so, I suggest clarifying that, e.g. "chemistry and ozone".

We modified the sentence in the following way: "understanding the physics and chemistry of sulfate stratospheric aerosols, their radiative impacts and other potential impacts on the stratosphere, e.g. on ozone".

p4, line 14: why do you expect the contribution of van der Waals forces to be insignificant? By changing the tails of the aerosol size distributions, van der Waals forces could possibly be significant over a long period of time, even if the impacts were negligible for a Pinatubo-size eruption. (see also my comment at p8, line 25).

As discussed above we have performed additional experiments including vdW enhancement and discuss them in the revised manuscript.

p4, line 26: How might prescribed oxidants impact results or introduce error? (do you expect the oxidant concentration to remain constant over a long period of time given continuous so2 injection?)

We expanded the paragraph by the following sentences: "For continuous SAI schemes, where the aerosol layer and its properties reach an equilibrium after several years, we would expect oxidants to reach equilibrium concentrations (somewhat) lower than the climatological values. Assuming a climatological chemical lifetime of SO_2 could therefore cause the model to overestimate SO_2 to H_2SO_4 conversion rates to a certain degree, favouring new particle formation over condensational growth."

p6, line 20: Change "completely" to something less exact, perhaps "essentially"

Done.

p8, line 25: The difference between your results and that of Niemeier and Timmreck (2015) is interesting. Can you estimate how much of the difference might be due to differences in the particle size distribution? (can you change your particle size distri- bution and run it through radiation code?) Other studies have found differences in the size distributions between modal models and sectional models. In an intercomparison study, Weisenstein et al. (2007) found sectional models to predict larger stratospheric aerosols after Pinatubo than modal models, and sectional models compared better to observations. Kokkola et al. (2009) completed an intercomparison between micro- physical and sectional versions of the ECHAM model and found significant differences in the evolution of stratospheric aerosol size distributions, with differences between modal and sectional models increasing with increasing SO2 injection rate. English et al. (2013) calculated aerosol mode widths from their sectional model assuming a log- normal distribution and found the mode width to change significantly after simulated volcanic eruptions, suggesting that 2-moment modal models with fixed mode widths may not accurately capture the evolving size distribution.

The are various differences between our model setup and that of Niemeier and Timmreck (2015) and it would be cumbersome to attribute the difference in results to the different factors. Going beyond the short analysis presented in the manuscript would require a dedicated intercomparison project such as GeoMIP. Differences in size distribution may indeed be a source of differences in the SW and LW RF but it is not appropriate to speculate based on global mean differences alone as geographical variation and details of the size distribution may matter. Differences in refractive indices may also explain some of the differences.

p9, line 32: Please cite Niemeier et al. (2011) who also studied injection height. The disagreement between your results and theirs is also interesting. Niemeier et al. (2011) also predicted increasing particle size with height (Fig 2b in their paper). Why might your results differ from theirs? Differences in LW radiation code? Differences in zonal transport with altitude? QBO?

Again answering these questions would require a significant amount of work and a dedicated intercomparison project with an agreed protocol. The model differences highlighted in our study may convince other modelling groups that such an intercomparison exercise is required in the future.

p10, line 13: Are you injecting only in 28 grid boxes? If so, the concentrations might still be high compared to other studies which continuously inject across all grid boxes spanning a latitude range (e.g. 30 S - 30 N). Also, please clarify the altitude for the Broad injection here.

Yes, in the BROAD scenario the injection take place in only 28 grid boxes. The injection height is the same as in the STANDARD scenario, i.e. 17$\pm$1 km everywhere.

p11, line 15: Please cite Aquila et al. (2012), who also quantified the impacts that aerosol radiative heating have on the peak injection altitudes (for Mount Pinatubo).

Done.

p13, line 22: How does this contradict English et al. (2013)? They did not mention radiative fluxes. Differences in AOD distribution might be because their model does not have aerosol radiative heating coupled.

Actually, we wanted to refer to English et al. (2012) instead of English et al. (2013). It is correct that English et al. (2012) only discuss the increase in aerosol burden and AOD for a higher injection altitude, not the resulting radiative forcing. We therefore removed the citation in this place.

p13, line 22: Cite Niemeier et al. (2011) in addition to Niemeier and Timmreck (2015). Again, why do your results differ if they also found an increase in particle radius with altitude?

We added a reference to Niemeier et al. (2011). It remains unclear why exactly they found an increase in radiative forcing for higher injection altitude despite the increase in particle size, while in our case the radiative forcing does not increase for higher injection altitudes.

p14, line 14: Expand this paragraph to further discuss the difficulties and complexities regarding geoengineered stratospheric aerosol (the processes are complex; the long lifetime allows for errors to amplifiy; geoengineering is only hypothetical and has no observations to evaluate models against (yet); potential uncertainties/errors in your model.

We modified the text in the following way: "The simulation results depend largely on the modelled evolution of particle size, radiative heating and stratospheric dynamics (controlling the spatial distribution). The stratospheric processes determining this are complex and uncertain and their implementation causes considerable disagreement between different models. In contrast to volcanic eruptions, models cannot be constrained by observations in the case of purely hypothetical geoengineering scenarios. Therefore, it would be worthwhile [...]".

Figure 4: the x-axis units label appears as "m" which is incorrect.

Figure 6: latitude axis shows all "0"

Figure 7d, 8, 10d, 11,12, and 14: y-axis values are cut off

We apologize for this. The figures got corrupted during the production process of the discussion paper and have already been corrected.

Figure 15: It would also be useful to have a plot of percent increase in deposition rate.

We agree that this would be an interesting information. But as our model (in the configuration used in this study with the S3A module) does not include deposition fluxes of tropospheric sulfate, it is not possible to compare the deposition of SAI sulfate to the deposition of tropospheric sulfate in detail.

Response to comments by Referee 3

This manuscript explore several geoengineering scenarios characterized by different injection amounts, altitudes, latitudes, and time of the year using the LMDZ-S3A model, which includes a sectional aerosol module. Several aspects of the simulations are here analyzed, both regarding the evolution of the aerosol and the effects on stratospheric dynamics.

Despite not being particularly innovative, this is an excellent paper that documents the extensive work performed by the author. It is a through analysis of different geoengineering scenarios within the same model framework, and the results are compared to previous publications when appropriate. I have only some very minor comments, mostly of technical nature.

We thank the reviewer for his/her general appreciation.

1 - Most figures were corrupted when generating the PDF (special characters and the scales of some axis are missing). Please correct them before publication.

We apologize for this. The figures got corrupted during the production process of the discussion paper and have already been corrected.

2 - Page 6 Line 24: I wonder if also the concentration (ug/m3) of sulfate peaks well above the injection height.

Figure 1 in this reply shows that the sulfate aerosol concentration in $\mu g\,S\,m^{-3}$ peaks at about $19\,km$, i.e. above the injection height of $17\,km$ but below the peak of the sulfate mass mixing ratio. We added this information to the text, but do not show the concentration plot in the revised manuscript.

3 - page 7 line 20: "The region with the largest particles descends towards higher latitudes due to ongoing particle growth and sedimentation during the meridional transport through the BDC." I am not sure I agree with this statement. This would imply that the effective radius at higher latitudes is larger than in the tropics, because particles have grown during the transport via BDC. The figure does not show that, though.

This is correct. We removed the words "due to ongoing particle growth" from this sentence.

4 - page 8 line 7: Figure 7 shows the absolute value of the forcing efficiency, correct?

No, the forcing efficiency is shown with positive and negative values, but the labels were corrupted in the previous version.

5 - page 9: Fig. 9 is not referred to in the test, right? I think it is a very useful figure that should be discuss. It can say something about how appropriate (or not) a modal approach is.

We added a reference to Figure 9 in the text.

6 - page 9 line 18: ", it appears that the net forcing/AOD decreases for higher injec- tions,". Am I looking at the orange line if Fig. 10b? That line increases with injection height.

This misunderstanding may have been caused by the corrupted axis labels. The negative value of the net forcing/AOD is approaching zero, i.e. its absolute value decreases with injection height.

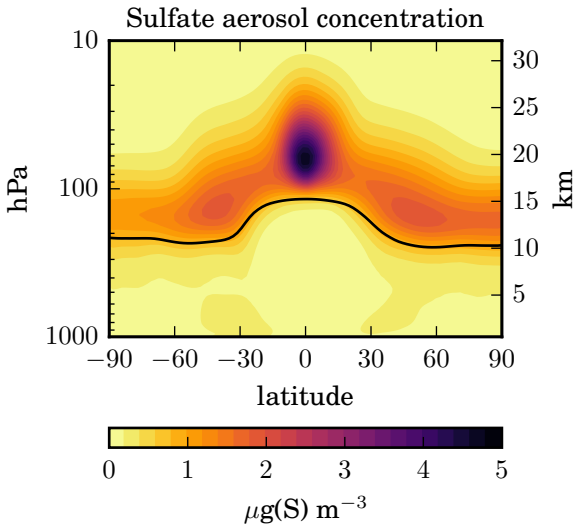

Figure 1. Latitude-height cross-section of the zonal and annual mean sulfate aerosol concentration (in $\mu g\,S\,kg\,m^{-3}$) for the STANDARD experiment.

7 - page 10 line 4: Are the authors referring to Fig. 12?

Yes, a reference to Figure 12 has been added.

8 - page 11 line 13: a difference plot would be useful, as the distributions are relatively similar.

Figure 2 in this reply shows the difference in aerosol extinction between the experiments. However, we decided to keep the absolute extinction plot in the manuscript.

Response to comments by Referee 4

The paper by Kleinschmitt et al., discusses the sensitivity of radiative forcing by stratospheric aerosol geoengineering to the injection amount and region using the LMDZ-S3A model and a section stratospheric aerosol model. The paper contributes to the question how efficient sulfur geoengineering can counteract radiative forcing. It is therefore an important contribution that should be published. The paper considers injection amounts as large as 50TgS to identify limits of this approach and also explores changes with regard injection locations. All experiments consider injections at the equator or within a region around the equator between 30N to 30S. In contrast to earlier studies, the amount of radiative forcing with injection amount is limited, which may be a result of the specific setup and the model. The paper is well written and I would recommend the publication to ACP after my comments and suggestions as listed below have been addressed.

We thank the reviewer for his/her positive comments.

General comment: One general comment is about the setup of the model. The use of fixed SSTs and non-interactive chemistry has limitations in correctly simulating the response of sulfur injections to dynamics. This has been mentioned in the manuscript,

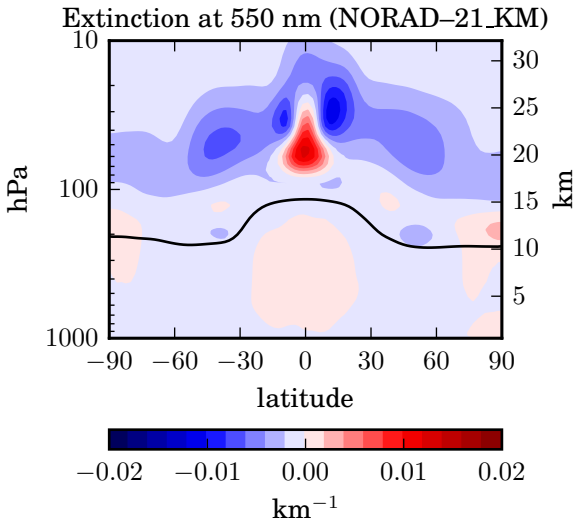

Figure 2. Latitude-height cross-section of the difference in the zonal and annual mean distribution of the aerosol extinction coefficient (km^{-1}) at 550 nm for equatorial stratospheric aerosol injections at 21 km between a simulation with non-radiatively interactive aerosol (NORAD) and a simulation with radiatively interactive aerosol (21_KM).

but a caveat along these lines needs to be also added in the discussion and conclusions of the paper.

We added the following paragraph to the conclusions: "We also have to keep in mind that our model setup does not include chemical feedbacks of the aerosol, e.g. on stratospheric ozone. Induced changes in ozone concentration might alter the radiative effects of SAI (especially the stratospheric heating) considerably. The prescription of sea surface temperatures constitutes another limitation for correctly simulating the feedback of SAI on atmospheric dynamics."

In addition, various figures in this paper do not show x and y axis annotations correctly.

We apologize for this. The figures got corrupted during the production process of the discussion paper and have already been corrected.

Specific comments:

Introduction: Line 18: SAI is a misleading abbreviation, aerosols are not injected. Please change to “sulfur injection” or “stratosphere aerosol geoengineering”

We agree that the term is not very precise, but we chose to keep it anyhow as it is widely used for the injection of aerosol precursors. We added the following sentence to the introduction: "Although we inject an aerosol precursor (i.e. SO_2) rather than sulfate aerosol, we loosely refer to the widely used term stratospheric aerosol injection (SAI) in this study."

Line 22: “using different aerosol types” is not correct. All the listed studies injected SO2 and some used H2SO4.

Ferraro et al. (2011) did study other aerosol types (titania, limestone, and soot). We believe the sentence is correct as it stands.

Page 3, Line 4: Can you give a reference that discusses the changes in the strength of the tropical pipe with regard to the phase

of the QBO?

We added a reference to Punge et al. (2009).

Line 23: What is pressure at the top of the model?

The top of the model is at a pressure of 4 Pa (i.e. about 80 km altitude). We added this information to the text.

Line 7: Is there a references that supports that commercial aircraft can hardly fly higher then 15km?

McClellan et al. (2012) state that "existing aircraft [...] are not optimized for high altitude flight and therefore are poorly suited to the geoengineering mission". But 15 km are not a hard limit, so we removed it from this sentence.

Line 10: Is there a reference that shows that injections at several locations may be more expensive and limited economies of scale? Why would injections at multiple locations be more effective? If there are studies discussing this, please mention those.

Of course this is somewhat speculative. We have changed the manuscript to say "There are few studies that look at the economic cost of SAI. However, we may naïvely expect that [...]"

Page 5, Line 15: The aerosol distribution is expected to adjust after 2 years, which needs to be shown. Even though 5 year simulations are rather short, I would recommend to average the last 3 years of the simulation to include the variability between the years. How will the budget in Figure 1 change if the last 3 years of the simulation are considered?

Figure 3 in this reply shows that the aerosol burden and size stabilise after 3 to 4 years and that the year to year variability is small. Therefore, we consider the results of the 5th year as representative. We added this information to the text.

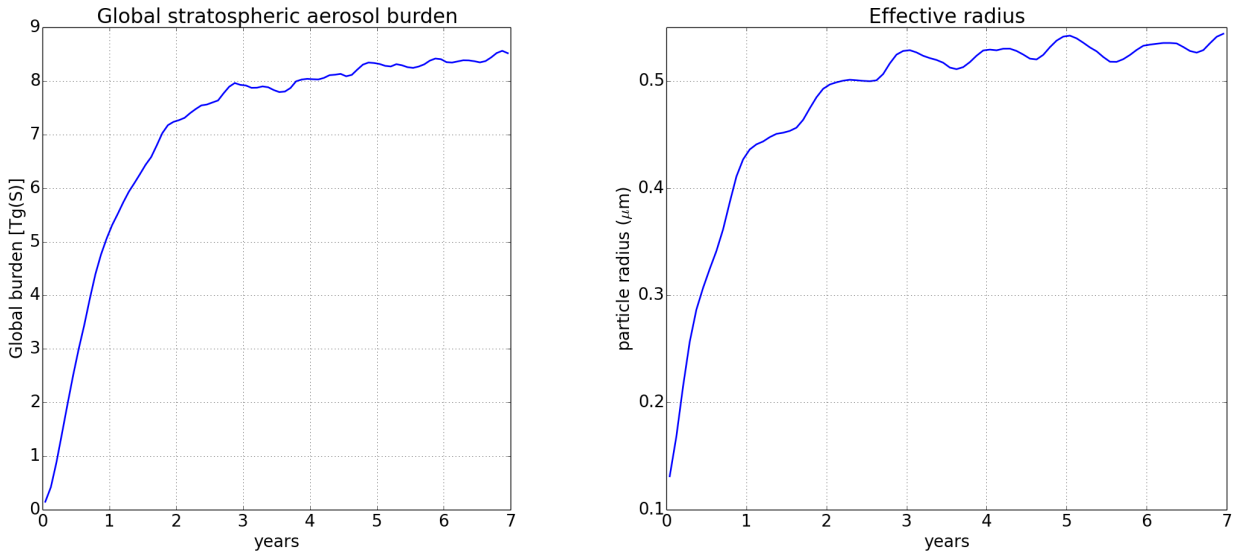

Figure 3. Time evolution of the global mean stratospheric sulfur burden (left, in Tg S) and sulfate aerosol effective radius (right, in µm) in the STANDARD experiment.

Page 7, Line 5: Total column ozone has decreased after volcanic eruptions in the past, but ozone mixing ratios do not decrease

everywhere in the stratosphere but also increase in the tropics due to the hydrolysis of N2O5 (Fahey et al., 1993).

We modified the sentence in the following way: "[...] whereas O_3 mixing ratios are known to have decreased in large parts of the stratosphere after Pinatubo, thus decreasing absorption of SW radiation. However, O_3 mixing ratios may also increase in the tropics due to the hydrolysis of N_2O_5 (Fahey et al., 1993), so the exact ozone feedback is unclear."

Page 9: 3.3 Sensitivity to injection height: what is the injection amount used in these sensitivity studies?

The injected amount in the experiments with different injection height is always the same as in the STANDARD experiment, i.e. $10\,\mathrm{Tg\,S\,yr^{-1}}$. We added this information to the first sentence of Sect. 3.3.

Page 13, Line 23: Niemeier and Schmidt (2017): is this study published in ACP?

The discussion paper has been online in ACPD since May 2017 and in the meantime the study has been accepted for publication in ACP. The reference was updated in the revised manuscript.

Page 14, last sentence: “and may be implicate stronger side-effects than initially thought”. What do you mean by this? What are the side effects you referring to?

We refer to the strong stratospheric heating and to its effects, i.e. the breakdown of the QBO and the rapid adjustments discussed in more detail in Boucher et al. (2017).

Figure 2, 3, 4, 6, 13: Latitudes not shown. What years of the simulation are shown?

The latitudes are shown in the corrected figures. As described in the text, all figures show results from the 5th year of the simulations, if not specified differently.

Figure 7, 8, 9: problems with y-axis

Done.

Figure 10, 11, 12: problems with y and x axis, and not clear what injection amount is shown.

Done.

Figure 15: how much is the amount compared to the tropospheric burden?

The numbers are compared in the text (page 12, line 4 and line 28).

Figure 16: color scale needs to be improved, very difficult to make out any structure.

We have improved the colour scale, but there is actually only little structure to see.

Summary of changes made in the revised manuscript

The changes in the manuscript resulting from the comments by the Referees are marked on the following pages.

References

Aquila, V., Oman, L. D., Stolarski, R. S., Colarco, P. R., and Newman, P. A.: Dispersion of the volcanic sulfate cloud from a Mount Pinatubo–like eruption, Journal of Geophysical Research: Atmospheres, 117, D06 216, doi:10.1029/2011JD016968, 2012.

Boucher, O., Kleinschmitt, C., and Myhre, G.: Quasi-additivity of the radiative effects of marine cloud brightening and stratospheric sulfate aerosol injection, Geophysical Research Letters, 44, 2017GL074 647, doi:10.1002/2017GL074647, in press, 2017.

English, J. M., Toon, O. B., and Mills, M. J.: Microphysical simulations of sulfur burdens from stratospheric sulfur geoengineering, Atmospheric Chemistry and Physics, 12, 4775–4793, doi:10.5194/acp-12-4775-2012, 2012.

English, J. M., Toon, O. B., and Mills, M. J.: Microphysical simulations of large volcanic eruptions: Pinatubo and Toba, Journal of Geophysical Research: Atmospheres, 118, 1880–1895, doi:10.1002/jgrd.50196, 2013.

Fahey, D. W., Kawa, S. R., Woodbridge, E. L., Tin, P., Wilson, J. C., Jonsson, H. H., Dye, J. E., Baumgardner, D., Borrmann, S., Toohey, D. W., Avallone, L. M., Proffitt, M. H., Margitan, J., Loewenstein, M., Podolske, J. R., Salawitch, R. J., Wofsy, S. C., Ko, M. K. W., Anderson, D. E., Schoeber, M. R., and Chan, K. R.: In situ measurements constraining the role of sulphate aerosols in mid-latitude ozone depletion, Nature, 363, 509–514, doi:10.1038/363509a0, 1993.

Ferraro, A. J., Highwood, E. J., and Charlton-Perez, A. J.: Stratospheric heating by potential geoengineering aerosols, Geophysical Research Letters, 38, L24 706, doi:10.1029/2011GL049761, 2011.

Kleinschmitt, C., Boucher, O., Bekki, S., Lott, F., and Platt, U.: The Sectional Stratospheric Sulfate Aerosol module (S3A-v1) within the LMDZ general circulation model: description and evaluation against stratospheric aerosol observations, Geoscientific Model Development, 10, 3359–3378, doi:10.5194/gmd-10-3359-2017, 2017.

Kokkola, H., Hommel, R., Kazil, J., Niemeier, U., Partanen, A.-I., Feichter, J., and Timmreck, C.: Aerosol microphysics modules in the framework of the ECHAM5 climate model – intercomparison under stratospheric conditions, Geoscientific Model Development, 2, 97–112, doi:10.5194/gmd-2-97-2009, 2009.

McClellan, J., Keith, D. W., and Apt, J.: Cost analysis of stratospheric albedo modification delivery systems, Environmental Research Letters, 7, 034 019, doi:10.1088/1748-9326/7/3/034019, 2012.

Niemeier, U. and Schmidt, H.: Changing transport processes in the stratosphere by radiative heating of sulfate aerosols, Atmospheric Chemistry and Physics Discussions, doi:10.5194/acp-2017-470, in press, 2017.

Niemeier, U. and Timmreck, C.: What is the limit of climate engineering by stratospheric injection of SO_2?, Atmospheric Chemistry and Physics, 15, 9129–9141, doi:10.5194/acp-15-9129-2015, 2015.

Niemeier, U., Schmidt, H., and Timmreck, C.: The dependency of geoengineered sulfate aerosol on the emission strategy, Atmospheric Science Letters, 12, 189–194, doi:10.1002/asl.304, 2011.

Pierce, J. R., Weisenstein, D. K., Heckendorn, P., Peter, T., and Keith, D. W.: Efficient formation of stratospheric aerosol for climate engineering by emission of condensible vapor from aircraft, Geophysical Research Letters, 37, L18 805, doi:10.1029/2010GL043975, 2010.

Punge, H. J., Konopka, P., Giorgetta, M. A., and Müller, R.: Effects of the quasi-biennial oscillation on low-latitude transport in the stratosphere derived from trajectory calculations, Journal of Geophysical Research: Atmospheres, 114, D03 102, doi:10.1029/2008JD010518, 2009.

Weisenstein, D. K., Penner, J. E., Herzog, M., and Liu, X.: Global 2-D intercomparison of sectional and modal aerosol modules, Atmospheric Chemistry and Physics, 7, 2339–2355, doi:10.5194/acp-7-2339-2007, 2007.

Sensitivity of the radiative forcing by stratospheric sulfur geoengineering to the amount and strategy of the SO_2 injection studied with the LMDZ-S3A model

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Abstract. The enhancement of the stratospheric sulfate aerosol layer has been proposed as a method of geoengineering to abate global warming. Previous modelling studies found that stratospheric aerosol injection could effectively compensate the warming by greenhouse gases on the global scale, but also that the achievable cooling effect per sulfur mass unit, i.e. the forcing efficiency, decreases with increasing injection rate. In this study we use the atmospheric general circulation model LMDZ with the sectional aerosol module S3A to determine how the forcing efficiency depends on the injected amount of SO_2, the injection height and the spatio-temporal pattern of injection. We find that the forcing efficiency may decrease more drastically for larger SO_2 injections than previously estimated. As a result, the net instantaneous radiative forcing does not exceed the limit of $-2\,\mathrm{W\,m^{-2}}$ for continuous equatorial SO_2 injections and it decreases (in absolute value) for ~~the largest injection rates simulated (50~~injection rates larger than $20\,\mathrm{Tg\,S\,yr^{-1}}$~~)~~. In contrast to other studies, the net radiative forcing in our experiments is fairly constant with injection height (in a range 17 to 23 km) for a given amount of SO_2 injected. Also spreading the SO_2 injections between 30°S and 30°N or injecting only seasonally from varying latitudes does not result in a significantly larger (i.e. more negative) radiative forcing. Other key characteristics of our simulations include a consequent stratospheric heating, caused by absorption of solar and infrared radiation by the aerosol, and changes in stratospheric dynamics, with a collapse of the quasi-biennial oscillation at larger injection rates, which has impacts on the resulting spatial aerosol distribution, size, and optical properties. But it has to be noted that the complexity and uncertainty of stratospheric processes cause considerable disagreement between different modelling studies of stratospheric aerosol geoengineering. This may be addressed through detailed model intercomparison activities, as observations to constrain the simulations of stratospheric aerosol injection are not available and analogues (such as volcanic eruptions) are imperfect.

1 Introduction

Solar radiation management (SRM) has been proposed as a possible means to offset increasing Earth's temperature in response to the anthropogenic greenhouse effect should mitigation efforts be insufficient and/or come too late. Among SRM techniques, stratospheric aerosol injection (SAI) has received a lot of attention, starting with Budyko (1977) and largely reinstated by Crutzen (2006). A large number of studies have been published since then, whose objectives fall in different categories : i)

understanding the ~~physico-chemistry~~ physics and chemistry of sulfate stratospheric aerosols, their radiative impacts and other potential impacts on the stratosphere, e.g. on ozone (e.g., Tilmes et al., 2008; Rasch et al., 2008; Pierce et al., 2010; Pitari et al., 2014; Aquila et al., 2014), ii) optimizing in some sense the stratospheric aerosol injection in space and time or by using different aerosol types (e.g., Heckendorn et al., 2009; Niemeier et al., 2011; Ferraro et al., 2011; English et al., 2012; Niemeier and Timmreck, 2015) , and iii) estimating the climate response to such stratospheric aerosol injection, and to which extent it compensates or not global warming expected from anthropogenic greenhouse effects (e.g., Robock et al., 2008; Tilmes et al., 2009; Ricke et al., 2010; Kravitz et al., 2013).

Despite an important research effort on SAI over the last 10 years, there remains a number of uncertainties and open questions. Recent reviews on the subject (e.g., Irvine et al., 2016; MacMartin et al., 2016; Visioni et al., 2017) highlighted the need for accurate stratospheric aerosol models. This is important indeed because the central idea of SAI is that stratospheric aerosols scatter sunlight back to space, thus cooling Earth, but they also absorb and emit infrared radiation, thus causing a warming influence. The net effect is sensitive to the aerosol spatial distribution and the aerosol microphysics, which in turn depend on an accurate representation of key processes in the stratosphere. In this study we make use of the new Sectional Stratospheric Sulfate Aerosol (S3A) module developed within the atmospheric general circulation model LMDZ, which is described in detail in Kleinschmitt et al. (2017).

We focus here on SRM using stratospheric sulfate aerosol (formed from injections of SO_2) because volcanic sulfate aerosol provides a natural analogue which largely confirms its capability to cool the Earth's surface. Other aerosol types have been proposed in the last years such as solid TiO_2, Al_2O_3, $CaCO_3$ or even diamond particles (Weisenstein et al., 2015; Dykema et al., 2016; Keith et al., 2016) because of their possibly more favourable optical and/or chemical properties. We do not explore these further in this article but our model could be adapted in the future to study other aerosol types or mixtures of different aerosol types. Although we inject an aerosol precursor (i.e. SO_2) rather than sulfate aerosol, we loosely refer to the widely used term stratospheric aerosol injection (SAI) in this study.

One open question regarding sulfate SAI is how the net radiative forcing scales with the amount of aerosol precursor or aerosol injected. At large sulfuric acid concentrations, particles tend to grow larger through condensation and coagulation, resulting in a decreased mass scattering efficiency for solar (shortwave = SW) radiation, while the mass absorption efficiency for terrestrial (longwave = LW) radiation remains essentially unchanged. This results in a less negative net forcing efficiency (i.e., radiative forcing per unit of injection rate) as the injection rate increases. Niemeier and Timmreck (2015) have studied the relation between sulfur dioxide injection rate and global mean radiative forcing in a series of simulations and found that for a 10-fold increase in injection rate (from 5 to 50 $\mathrm{Tg\,S\,yr^{-1}}$), the net forcing efficiency is reduced by 50 %.

The competition between the (mostly negative) SW radiative forcing and (mostly positive) LW radiative forcing of stratospheric aerosols is an important issue, as both effects can be of similar magnitude. As the net forcing is the sum of these partly compensating terms which both have considerable uncertainty, the overall uncertainty of the net forcing is even larger. For this reason, we discuss SW, LW and net (SW+LW) forcings independently in this study. Here, we focus on instantaneous radiative forcings, but the radiative effects of rapid adjustments (leading to an effective radiative forcing) are discussed in a companion article (Boucher et al., 2017) in the context of combining stratospheric aerosol injection and marine cloud brightening.

Another important parameter of SAI is the injection height for the aerosol or its precursors. In principle, one would expect the forcing efficiency to increase with injection height, because it would take more time for sedimentation to transport the aerosol below the tropopause. However sedimentation is not the only relevant process. Stratospheric dynamics also plays an important role in determining the aerosol spatial distribution and therefore its lifetime. Aerosol injected or formed in the lower stratosphere may be transported polewards and return into the troposphere quicker than aerosol injected in middle stratosphere because the lower branch of the meridional Brewer-Dobson circulation is more rapid than the higher branch. For aerosols injected in the tropics, the speed of the meridional transport also depends on the phase of the Quasi-Biennial Oscillation (QBO) (Punge et al., 2009) . During the QBO easterly phase, the subtropical barrier is more effective and the tropical pipe is stronger, thus, the meridional transport is slower. In contrast, during the QBO westerly phase the subtropical barrier is less effective, the tropical pipe is shifted up, and the meridional transport is faster. However it could be that large aerosol injections feed back on stratospheric circulation and hence on the aerosol stratospheric lifetime (Aquila et al., 2014; Niemeier and Schmidt, 2017).

At the same time, the "effort" spent on SAI and thus the cost of SAI measures strongly depends on injection height, because conventional aircraft ~~can hardly fly higher than 15~~are not optimised for flying at high altitude. Therefore, the development of a special aircraft fleet or an alternative technology aimed at higher injections, if required, would result in significantly higher economic costs (McClellan et al., 2012). The costs as a function of injection height are nevertheless not well established, so it is not straightforward to decide if it is cheaper to inject a larger quantity at a lower altitude or a smaller quantity at a higher altitude. ~~Likewise~~ There are few studies that look at the economic cost of SAI. However, we may naïvely expect that the injection at several locations may be more expensive (because it would require more infrastructure and limit economies of scale) or less expensive (because it would be more effective to create radiative forcing). Nevertheless, the objective of this study is not to find the cheapest injection strategy but to investigate how the radiative forcing depends on the injected amount of sulfur, the injection height, the number of injection points, and the temporal pattern of the injection. In this context we also study the impact of SAI on stratospheric dynamics and to which extent the additional aerosol of stratospheric origin increases particle concentrations and acid deposition at the Earth's surface.

2 Model and simulations

2.1 Model description

In this study we use the atmospheric general circulation model LMDZ (Hourdin et al., 2006, 2013) with the new sectional stratospheric sulfate aerosol module S3A described in detail in Kleinschmitt et al. (2017).

In the present configuration, LMDZ is run with 96×96 grid-points on the horizontal – i.e., a resolution of 1.89° in latitude and 3.75° in longitude which is the same as for LMDZ5A (Hourdin et al., 2006, 2013) – and 79 layers on the vertical in order to improve the vertical resolution in the stratosphere. The pressure at the top of the model is 4 Pa (corresponding to roughly 80 km altitude). Although it is possible to couple LMDZ interactively to an ocean model to form the IPSL coupled atmosphere-ocean model, we decided here to only perform simulations with fixed sea surface temperatures (SST). This is because our focus is on estimating radiative forcings and because this choice allows us to perform more and longer simulations for a given computa-

tional cost. It has to be highlighted that due to the fixed SST it is not possible to evaluate the overall impact of SAI on climate, but we can analyse instantaneous radiative effects of the aerosol as well as the fate of the sulfate particles in the troposphere. Rapid adjustments on the temperature, water vapour and cloud fields are discussed in a separate study (Boucher et al., 2017).

Special attention was paid to the simulation of stratospheric dynamics in the model in general, and the Brewer-Dobson circulation and the quasi-biennial oscillation (QBO) in particular (de la Cámara et al., 2016; Kleinschmitt et al., 2017). Since the relevance of certain physical processes varies between the troposphere and the stratosphere, it is important to diagnose the tropopause during the simulation. Therefore, LMDZ-S3A computes the tropopause pressure at every time step following the WMO definition based on the lapse rate (Reichler et al., 2003). The tropopause height is used to estimate the budget of stratospheric aerosols.

S3A includes a representation of sulfate particles with dry radii between 1 nm and 3.3 µm discretised in 36 logarithmically spaced size bins, as well as the precursor gases OCS, SO_2 and H_2SO_4. The physical processes of nucleation, condensation, evaporation, coagulation and sedimentation are represented. It should be noted that we only consider coagulation of aerosol particles from Brownian diffusion (Jacobson et al., 1994) and do not account for the enhancement effect due to van der Waals (vdW) forces when computing the coagulation kernels. It has been shown that van der Waals forces may enhance coagulation by 30% for the smallest particles but their importance decreases rapidly as particle size increases (Chan and Mozurkewich, 2001). Sensitivity studies performed by English et al. (2013) and Sekiya et al. (2016) simulating the 1991 eruption of Mount Pinatubo found that including inter-particle van der Waals forces increased the peak effective radius by 10 % and decreased stratospheric AOD and burden by 10 %. A further sensitivity test performed in our model has shown that considering the van der Waals coagulation term resulted in a relatively small shift in the particle size distribution towards larger sizes and a small decrease in the simulated peak global-mean stratospheric aerosol optical depth in the weeks following the Pinatubo eruption (Kleinschmitt et al., 2017). ~~We expect this effect to be smaller for our continuous SAI experiments and we neglect it here given~~ Given the large uncertainties on how to parametrise ~~it~~the vdW effect on coagulation, we decided not to consider this process in most of our experiments. However, in Sect. 3.5 we discuss the results of a sensitivity test including vdW coagulation enhancement under conditions of strong SAI.

An important feature of our model is the fact that aerosols are fully interactive with the radiative transfer scheme, thus allowing changes in atmospheric heating rates due to stratospheric aerosols. We showed in Kleinschmitt et al. (2017) that aerosol heating causes an upward motion that lifts the aerosol in a simulation of the Pinatubo eruption. Aerosol optical properties are computed from Mie theory using the simulated size distribution and a prescribed chemical composition consisting of 75% H_2SO_4. We compute spectrally-averaged optical properties for the 6 solar and 16 terrestrial wavebands of the radiative transfer model as well as for the 550 nm and 10 µm wavelengths for the sake of analysing the results.

A limitation of our experimental setup is the assumed fixed chemical timescale for SO_2 to H_2SO_4 conversion. This neglects any feedbacks of OH depletion onto SO_2 oxidation rate and lifetime. While this effect has been shown to be important for exceptional eruptions such as that of Tambora (Bekki, 1995), we estimated that this effect only limits oxidation for a short period after an eruption of the size of Pinatubo (Kleinschmitt et al., 2017) with simulated maximum grid-box SO_2 mass mixing ratios of 71 ppm (in daily mean) and 2.6 ppm (in monthly mean). We therefore expect this effect to be smaller for our continuous

Table 1. Description of the experiments performed in this study.

Experiment	Description
CONTROL	Control experiment with background stratospheric sulfate aerosols
STANDARD	Continuous equatorial injection of 10 $\mathrm{Tg\,S\,yr^{-1}}$ as SO_2 at 17±1 km into one equatorial grid cell
x_TGS	Same as STANDARD, but with different injection rates, where x = 2, 5, 20, 50 $\mathrm{Tg\,S\,yr^{-1}}$
z_KM	Same as STANDARD, but with different injection heights, where z = 15, 19, 21, 23 km
BROAD	Same as STANDARD, but injection distributed at 28 locations around the globe between 30°N and 30°S
SEASONAL	Same as STANDARD, but injections into one grid cell limited to two months of the year (at 5°N in April and at 5°S in October)
x_TGS, $E(0)/E(\infty)$	Same as x_TGS experiments (with x = 20, 50 $\mathrm{Tg\,S\,yr^{-1}}$), but with coagulation enhanced by van der Waals forces (continuum regime factor $E(0)$ and kinetic regime factor $E(\infty)$, respectively)
NORAD	Same as STANDARD, but with radiatively non-interactive aerosol and an injection height of 21 km

SAI experiments where maximum SO_2 mixing ratios do not exceed 3.4 ppm in daily mean (and 2.2 ppm in monthly mean) for a continuous injection rate of 10 $\mathrm{Tg\,S\,yr^{-1}}$ and 16 ppm in daily mean (8.4 ppm in monthly mean) for 50 $\mathrm{Tg\,S\,yr^{-1}}$. For continuous SAI schemes, where the aerosol layer and its properties reach an equilibrium after several years, we would expect oxidants to reach equilibrium concentrations (somewhat) lower than the climatological values. Assuming a climatological chemical lifetime of SO_2 could therefore cause the model to overestimate SO_2 to H_2SO_4 conversion rates to a certain degree, favouring new particle formation over condensational growth.

2.2 Simulation setup

We define a STANDARD SAI scenario, upon which we then base sensitivity studies to estimate the role of certain parameters and features of the injection strategy. In this standard scenario, an amount of 10 $\mathrm{Tg\,S\,yr^{-1}}$ is injected in the form of SO_2 gas into one equatorial grid cell (at 1°N, 120°E). The equatorial injection takes advantage of the ascending branch of the Brewer-Dobson circulation (BDC) that contributes to lift the aerosol. The aerosol lifetime in the stratosphere in this configuration can be expected to be longer than for injection at higher latitudes, because of the poleward transport and subsequent removal through the BDC. Furthermore, the radiative forcing of the aerosol is generally larger (i.e., more negative) in the tropics because of the maximum in insolation. The injection is constant in time, i.e. we add the same amount of SO_2 at every single time step of 30 min. Vertically the injection is distributed with a Gaussian profile in altitude centred at 17 km with a standard deviation of 1 km (similarly to our setup of the 1991 eruption of Mount Pinatubo discussed in Kleinschmitt et al. (2017)).

In a first series of model experiments the magnitude of the stratospheric injection is varied between 2 and 50 $\mathrm{Tg\,S\,yr^{-1}}$, while all the other parameters are kept constant (experiments labelled x_TGS, see Table 1). In a second series the central injection height is varied between 15 and 21 km (still with a standard deviation of 1 km) for a default injection rate of 10 $\mathrm{Tg\,S\,yr^{-1}}$ (experiments labelled z_KM).

In addition to the sensitivity studies for injection magnitude and height, we performed experiments with different spatio-temporal injection patterns (BROAD and SEASONAL). In BROAD, the injection is spread equally in 28 locations with 7 latitudes (30°S, 20°S, 10°S, 1°N, 10°N, 20°N, 30°N) and 4 longitudes (120°W, 30°W, 60°E, 150°E). In SEASONAL, the injection is performed in the months of April and October only, switching from a location at 5°N to a location at 5°S, so as to always emit from the summer hemisphere. The STANDARD, BROAD and SEASONAL simulations therefore represent different injection strategies in terms of locations and logistics.

Each simulation is performed for 5 years and the results are shown for the 5th year of the simulation, when the aerosol layer has clearly reached a steady state (with only little year to year variability) regarding burden, particle size and spatial distribution. The stratospheric model fields are generally averaged over all the grid cells above the tropopause (diagnosed at the grid cell and timestep resolution).

We define the radiative forcing as the all-sky aerosol direct radiative effect (ADE) at the top of the atmosphere (TOA) in $\mathrm{W\,m^{-2}}$ for shortwave (SW), longwave (LW) and net radiation. At each time step the radiative forcings are computed as the difference of two calls of the radiation code (one including sulfate aerosol, one not). Therefore the radiative forcing does not include any rapid adjustments that take place in the land-atmosphere system, including changes in stratospheric temperature and water vapour. The differences between (instantaneous) radiative forcing and effective radiative forcing (ERF) that include rapid adjustments are discussed in a separate study (Boucher et al., 2017). It should also be noted that the change in stratospheric water vapour may also feedback onto stratospheric chemistry and SO_2 oxidation, which is not accounted for in our simulations. We have to point out that the presented radiative forcing values from the SAI model experiments include a small contribution from the background sulfate aerosol, which we do not subtract for simplicity. In the control experiment, the background sulfate aerosol has a global and annual mean net instantaneous radiative forcing of –0.04 $\mathrm{W\,m^{-2}}$ (–0.05 $\mathrm{W\,m^{-2}}$ in the SW and +0.01 $\mathrm{W\,m^{-2}}$ in the LW).

In order to get a better understanding of the relation between forcing efficiency (in $\mathrm{W\,m^{-2}}$ / $\mathrm{Tg\,S\,yr^{-1}}$) and injection rate magnitude, we decompose it in the following way:

$$\text{forcing efficiency} = \frac{\text{radiative forcing}}{\text{injection rate}} = \frac{\text{radiative forcing}}{\mathrm{AOD}(550\,\mathrm{nm})} \cdot \frac{\mathrm{AOD}(550\,\mathrm{nm})}{\text{burden}} \cdot \frac{\text{burden}}{\text{injection rate}} \tag{1}$$

The first term (radiative forcing/AOD(550 nm) in $\mathrm{W\,m^{-2}}$ / unit AOD) reflects how a certain optical depth translates into a radiative forcing and is related to the model's radiative transfer code, to the distribution of the stratospheric aerosols in space and time, but also to particle size (since for a given AOD(550 nm), larger particles have a larger LW forcing than small particles). The second term (AOD(550 nm)/burden in unit AOD / $\mathrm{Tg\,S}$) shows how effectively the particles extinguish visible light and is related to particle size (and hence to the model's aerosol microphysics). The third term (burden/injection rate in $\mathrm{Tg\,S}$ / $\mathrm{Tg\,S\,yr^{-1}}$ = yr) is equal to the effective lifetime of the injected sulfur in the stratosphere and therefore strongly depends on the modelled transport and removal processes. It should be noted that this decomposition is similar to the one used by Schulz et al. (2006) but is applied here to stratospheric aerosols.

3 Simulation results

3.1 Results from the STANDARD experiment

Figure 1 shows the stratospheric burdens and the fluxes of the represented sulfur species for the STANDARD SAI simulation with an equatorial injection of $10\,\mathrm{Tg\,S\,yr^{-1}}$ at $17\,\mathrm{km}$ altitude. The injected SO_2 is converted to H_2SO_4 after a mean lifetime of 41 days, while a fraction of roughly 4 % leaves the atmosphere without being converted (mostly because it is injected below the tropopause and rapidly removed from the troposphere through wet scavenging). Conversion of OCS to SO_2 in the stratosphere (after a mean lifetime of 8 years) adds only 0.3 % to the injected sulfur mass. Therefore the stratospheric sulfur budget can be considered as ~~completely~~ essentially anthropogenic in this scenario. H_2SO_4 vapour has a lifetime of 2 days, with the nucleation and condensation processes converting H_2SO_4 vapour to the particulate phase. Thus 85 % of the stratospheric sulfur exists in the form of H_2SO_4 aerosols which has a mean lifetime in the stratosphere of about 10 months.

The spatial distribution of the sulfate aerosol mass mixing ratio (in $\mu\mathrm{g\,S\,kg\,air^{-1}}$) in Fig. 2 shows a pronounced maximum in the tropical stratosphere around $21\pm4\,\mathrm{km}$ altitude, i.e. well above the injection zone at $17\pm1\,\mathrm{km}$. The sulfate concentration (in $\mu\mathrm{g\,S\,m^{-3}}$, not shown) peaks at about $19\pm3\,\mathrm{km}$, i.e. above the injection height but below the peak of the sulfate mass mixing ratio. Outside the tropics the sulfate mixing ratio is much lower, characteristic of a relatively strong tropical transport barrier, and decreases further towards the poles. The spatial distribution of the injected SO_2 (not shown) is much more confined to the latitudinal band corresponding to the injection zone although it expands vertically a few km above the injection altitude. This is because the injected SO_2 is almost completely converted to sulfuric acid gas before significant latitudinal transport to higher latitudes can take place. The mixing ratio of sulfuric acid vapour (not shown either) is very low in the lower stratosphere due to its rapid consumption through particle formation and growth, but it increases above $30\,\mathrm{km}$, where the aerosol starts to evaporate.

The absorption of SW and LW radiation by the aerosol can heat up the surrounding air considerably. The temperature anomaly in the STANDARD SAI scenario with respect to the CONTROL run is shown in Fig. 3. It reaches up to $16\,\mathrm{K}$ in the lower tropical stratosphere and decreases with increasing altitude and latitude. In the upper troposphere the heating is weaker due to the low particle concentrations, but it can reach up to $3\,\mathrm{K}$ just below the tropopause. This heating is significant and contributes to the lifting of the injected SO_2 and the aerosol. We evaluated this heating for the Pinatubo simulation performed in Kleinschmitt et al. (2017) and found that the model may overestimate the temperature anomaly by a factor of 2 relative to MSU channel 4 observations. One possible reason for the discrepancy was that we prescribed O_3 to its climatological values, whereas O_3 mixing ratios are known to have decreased in large parts of the stratosphere after Pinatubo, thus decreasing absorption of SW radiation. However, O_3 mixing ratios may also increase in the tropics due to the hydrolysis of N_2O_5 (Fahey et al., 1993) , so the exact ozone feedback is unclear. Therefore, the effects of SAI presented in this study that critically depend on the radiative heating may not be as important in reality and should be studied with other models as well.

The tropopause itself, which is diagnosed within the model following the WMO definition based on the lapse-rate, descends by about $1\,\mathrm{km}$ in the tropics due to the heating of the lower stratosphere and the upper troposphere. Santer et al. (2003) studied the contribution of various forcings to observed tropopause height changes. They showed that after the Mount Pinatubo eruption a

tropopause descent of 5–10 hPa (roughly 0.2–0.4 km) was observed. This was probably caused by a heating of the stratosphere and a parallel cooling of the troposphere by the volcanic sulfate aerosol. But the underlying trend over the last decades is an increase in tropopause height caused by well-mixed greenhouse gases, which heat the troposphere and cool the stratosphere. Hence, this observed increase due to global warming might be compensated (or even overcompensated) by the effect of SAI. But it should also be noted that our simulations are based on fixed SST, while an interactive ocean might have an impact on temperature changes and therefore on the tropopause height.

The spatial distribution of particle size (i.e., effective radius r_{eff}) is shown in Fig. 4. With effective radii up to 0.5 $\mu\mathrm{m}$ the particles are largest above the injection region where the sulfate concentrations are the largest. The region with the largest particles descends towards higher latitudes due to ongoing particle growth and sedimentation during the meridional transport through the BDC.

3.2 Sensitivity to injection magnitude

The relation between the magnitude of the stratospheric sulfur injections and the resulting net radiative forcing is shown in Fig. 5. The most surprising observation is that the forcing does not exceed the 1.9 $\mathrm{W\,m^{-2}}$, which is reached in the 20_TGS experiment, but decreases for an even larger injection rate of 50 $\mathrm{Tg\,S\,yr^{-1}}$. This is because the SW forcing does not increase as strongly with injection magnitude as the LW forcing (with opposite sign), due to the increase in particle size.

In order to illustrate the non-linear impact of the injection magnitude on the spatial distribution of the stratospheric aerosol, we plotted the sulfate aerosol mass mixing ratio (MMR) normalised by the value in the STANDARD experiment and the ratio of the injection rates in Fig. 6. For a linear scaling of MMR with injection rate, this ratio would be unity everywhere (so the base-10 logarithm of the ratio –which is the quantity plotted– would be zero). In our model simulations, for a small injection of 2 $\mathrm{Tg\,S\,yr^{-1}}$, the ratio is well below 1 above and below the injection region, while it is above 1 in the polar stratosphere. For a large injection of 50 $\mathrm{Tg\,S\,yr^{-1}}$, the ratio shows the opposite pattern (apart from the zone immediately above the injection region). From this we can conclude that in relative terms with increasing magnitude of the injection rate 1) more sulfur accumulates below the injection region because of increasing sedimentation of larger particles and 2) less sulfur reaches the high latitudes because of the induced changes in stratospheric dynamics (discussed in Sect. 3.7) and/or because of faster sedimentation during the meridional transport. The sulfate MMR above the injection region is largest (in relative terms) in the 10 and 20 $\mathrm{Tg\,S\,yr^{-1}}$ scenario, probably because in these cases the updraft by aerosol heating is strong enough to lift the particles while the sedimentation is still relatively slow. The mean forcing efficiency and its decomposition described above for the simulations with varying injection magnitude are shown in Fig. 7. The forcing efficiency in the SW decreases by roughly 50 % between 2 and 50 $\mathrm{Tg\,S\,yr^{-1}}$, while in the LW it stays rather constant. As a result, the net forcing efficiency decreases dramatically for larger injection rates (by 94 % between 2 and 50 $\mathrm{Tg\,S\,yr^{-1}}$). Even the absolute forcing decreases from 20 to 50 $\mathrm{Tg\,S\,yr^{-1}}$ (see Fig. 5). The decomposition shows that this decrease is mainly due to a decreasing net forcing/AOD (by 87 % between 2 and 50 $\mathrm{Tg\,S\,yr^{-1}}$ injection rates), which itself is caused by an increasing LW forcing/AOD combined with a rather constant SW forcing/AOD. The AOD/burden also decreases by more than 50 % due to larger particles (see Fig. 8). Despite this finding, the lifetime of the particles does not clearly decrease for larger injection magnitude. This may be explained by the

superposition of two opposed effects: increasing sedimentation velocity and increasing updraft through heating of the air. Both can be seen in Fig. 6a, where the sulfate mixing ratio increases disproportionately with the injection rate above the injection region (due to updraft) and below it (due to sedimentation). But for the largest injection rate simulated, enhanced sedimentation starts to dominate (see Fig. 6b) so that the particle lifetime can be expected to decrease further for even larger injections.

Figure 8 shows the impact of the injection magnitude on particle size. The global mean effective radius (computed for all the aerosol above the tropopause) almost triples between 2 and 50 $\mathrm{Tg\,S\,yr^{-1}}$. The mass size distribution shown in Fig. 9 mainly differs in the size range above 0.1 $\mu\mathrm{m}$ such that the mode radius grows and is shifted towards larger radius with increasing injection rate. This explains the increase in LW forcing and therefore the decreasing forcing efficiency.

Comparison with results from Niemeier and Timmreck (2015)

Niemeier and Timmreck (2015) performed a similar sensitivity study for the SAI injection rate with the ECHAM model using a modal aerosol module. Their results deviate from ours in that the absolute net forcing increases ~~monotonously~~ monotonically with injection rate up to 100 $\mathrm{Tg\,S\,yr^{-1}}$, while the forcing efficiency decreases only moderately (see crosses in Fig. 7).

One important difference in the results of the two models is the evolution of particle size with increasing injection rate. Fig. 8 shows that in LMDZ-S3A the particles are smaller than in ECHAM for small injections and larger for large injections. Therefore the positive LW forcing increases more in LMDZ-S3A, resulting in a lower net forcing efficiency. This difference in particle growth may partly be caused by differences in aerosol microphysics (modal versus bin scheme) and/or differences in meridional transport between the two models. The transport barrier is quite weak in the ECHAM version with 39 vertical levels (without a generated QBO) used by Niemeier and Timmreck (2015) and transport is hardly affected by the aerosol, while in LMDZ-S3A the barrier is strengthened for larger injection rates because of the impact of the radiative heating on the stratospheric dynamics. Therefore the aerosol plume is even more confined for larger injections, leading to enhanced particle growth. In a recent study, Niemeier and Schmidt (2017) used a version of ECHAM with higher vertical resolution (L90) and more realistic stratospheric dynamics and also found that heating by aerosols slows down the meridional transport, making larger injections even less efficient.

We also found that the maximum extinction coefficient (in $\mathrm{km^{-1}}$) in both the SW and the LW bands is about a factor of 2 larger in LMDZ-S3A for a given injection rate. This causes a stratospheric heating which is also about twice as strong as in ECHAM. The stronger heating may explain why aerosol lofting plays a more important role in LMDZ-S3A, such that increasing sedimentation can be partly compensated (resulting in a longer aerosol lifetime). The difference in extinction between the models might be due to different particle size distributions, different computations of the aerosol optical properties or due to differences in the radiative transfer scheme.

Another difference may also partly explain the differing model results: In ECHAM, the global mean SW forcing/AOD increases for larger injections, keeping the net forcing/AOD rather constant. In contrast, in LMDZ-S3A the SW forcing/AOD is rather independent of the injection rate, which in combination with increasing LW forcing/AOD causes the net forcing/AOD to decrease drastically. It remains unclear why the SW forcing/AOD increases with injection rate in ECHAM.

3.3 Sensitivity to injection height

Earlier model studies found a strong dependence of the forcing efficiency on the injection height, therefore we conducted a series of simulations with different injection heights (but all with the same injection rate of 10 $\mathrm{Tg\,S\,yr^{-1}}$).

Figure 10 shows that both the SW and the LW forcings increase with increasing injection height, but the net forcing (sum of SW and LW components) is almost completely independent of the injection height. Considering again the decomposition of the forcing efficiency given in Eq. 1, it appears that the net forcing/AOD decreases for higher injections, implying a less optimal size and spatial distribution of the aerosol. The AOD/burden ratio decreases as well due to larger particles (see Fig. 11), while the lifetime increases, as one would expect. There are various reasons for a shorter aerosol lifetime in the stratosphere for lower injection height:

- A larger fraction of the sulfur is injected below the tropopause, so most of it does not enter the stratosphere at all and is rapidly removed from the troposphere. In the 15_KM experiment 72 % of the sulfur are injected below the tropopause, while in the 17_KM (STANDARD) experiment it is only 4 %.

- In the lower stratosphere the tropical meridional transport barrier is less pronounced than above 20 km. Therefore the sulfur is transported more effectively to higher latitudes, where it leaves the stratosphere again.

The lifetime increases for higher injections, but the mean effective particle radius increases as well. Particles might grow larger because the aerosol does not spread as rapidly as in the lower stratosphere and the larger local concentrations favour coagulation and condensation. Additionally, in the case of higher injections the mean stratospheric size distribution contains more particles which had more time to grow due to their longer lifetime.
The effects of longer lifetime and less optimal optical properties (larger particles) just cancel out each other, such that the overall radiative forcing does not increase with injection height. As higher injections can be expected to be technically challenging, the effort to produce a certain forcing even increases. Therefore our modelling results imply that it would not be worthwhile to inject the aerosol at altitudes higher than 19 km. However the simulations show that injections at less than 17 km altitude would probably be very inefficient, because they result in a small stratospheric aerosol burden.

The particle size, i.e. the effective radius, shown in Fig. 11 appears to approach a saturation level of slightly below 0.5 µm with increasing injection height. Only the aerosol mass on the lower end of the size range decreases further (see Fig. 12), probably because of faster coagulation. The particle concentration in all the size bins decreases when going from the tropics to mid and high latitudes, except for the 15_KM experiment, where the concentration of particles around 0.1 µm slightly increases towards mid latitudes. This may be due to relatively fast meridional transport of the aerosol through the lower branch of the BDC in this case.

Table 2. Global mean quantities for experiments with 10 Tg S yr^{-1} injection rate but different spatio-temporal injection patterns.

Experiment	Radiative Forcing ($W\,m^{-2}$)			AOD at 550 nm	r_{eff} (μm)	~~burden~~ Aerosol Burden (Tg S)
	SW	LW	net			
STANDARD	–4.6	+3.1	–1.5	0.19	0.37	8.1
BROAD	–3.7	+2.2	–1.5	0.16	0.30	5.7
SEASONAL	–4.8	+3.1	–1.6	0.20	0.38	8.4

3.4 Sensitivity to spatio-temporal injection pattern

Previous model studies (e.g., Niemeier and Timmreck, 2015) indicate that the choice, when and where to inject a given mass of SO_2, can affect the forcing efficiency considerably. Therefore we simulated two additional scenarios with an injection rate of 10 Tg S yr^{-1}, but with injection patterns differing in time and space.

In the BROAD scenario, in which the sulfur mass is distributed over 28 points covering a larger area, the effective particle radius r_{eff} is almost 20 % below the one in the STANDARD scenario with injection into one equatorial grid box. However, the net forcing is almost equal in the two experiments, because the more favourable optical properties in the BROAD simulation are compensated by a smaller stratospheric aerosol burden. One reason for the smaller burden (and shorter lifetime) is the shorter average travel time from the injection region (30°N to 30°S) to mid/high latitudes where the aerosol is removed from the stratosphere. Another cause is that most of the aerosol remains below 20 km altitude, because it is not lifted by the BDC at the Equator and the more distributed injection pattern produces smaller radiative heating rates and therefore less updraft than in the STANDARD scenario. The AOD at 550 nm is much smaller in the tropics, but larger at mid latitudes.

In the SEASONAL scenario, where the sulfur mass is wholly injected during only two months of the year, the particles are slightly larger than in the STANDARD scenario. But because the burden is also a bit larger, the resulting net forcing is about 10 % larger than in the STANDARD scenario. The larger burden may result from a stronger updraft (due to larger aerosol concentrations in the short injection periods) that transports the aerosol to higher altitudes. This small increase in forcing efficiency seems to contradict a recent study by Laakso et al. (2017) who did not find a larger SW radiative forcing for seasonally varying injection scenarios. But it may be that the increase that we find is only caused by the stronger updraft / longer lifetime and not by the seasonal variations.

3.5 Sensitivity to the van der Waals coagulation enhancement factor

As argued in Sect. 2.1, the used model configuration of LMDZ-S3A does not include the enhancement of coagulation by van der Waals (vdW) forces, because sensitivity tests under Pinatubo conditions (Kleinschmitt et al., 2017) did not clearly improve the agreement with aerosol observations and there are large uncertainties on how this effect should be parametrised. In order to test if neglecting vdW forces is also justified under conditions of strong SAI, we performed additional sensitivity tests by repeating the 20_TGS and the 50_TGS experiments, once with the continuum regime vdW enhancement factor $E(0)$ and once

Table 3. Global mean quantities for experiments with and without van der Waals (vdW) enhancement of coagulation at injection rates of 20 and 50 Tg S yr^{-1}, respectively. The enhancement factor $E(\infty)$ for the kinetic regime is generally larger than the factor $E(0)$ for the continuum regime. Uniformly enhancing coagulation by $E(\infty)$ can therefore be seen as an upper limit of the impact of vdW forces.

Experiment	Radiative Forcing (W m^{-2})			AOD at 550 nm	r_{eff} (µm)	Aerosol Burden (Tg S)
	SW	LW	net			
20_TGS, no vdW	–8.1	+6.3	–1.9	0.33	0.46	17.1
20_TGS, $E(0)$	–7.7	+6.0	–1.7	0.31	0.47	16.2
20_TGS, $E(\infty)$	–6.4	+5.5	–0.9	0.24	0.52	14.0
50_TGS, no vdW	–14.8	+13.7	–1.0	0.62	0.60	41.1
50_TGS, $E(0)$	–14.1	+13.5	–0.6	0.57	0.63	40.0
50_TGS, $E(\infty)$	–11.9	+12.2	+0.4	0.45	0.67	34.4

with the (generally larger) kinetic regime vdW enhancement factor $E(\infty)$ from Chan and Mozurkewich (2001) . The actual enhancement factor for stratospheric conditions can be expected to lie in between these two cases.

Figure 13 shows that including a vdW enhancement factor causes a small shift in the aerosol size distribution towards larger radii. The effect is stronger for larger injection rates and for the larger enhancement factor $E(\infty)$, so that e.g. in the 50_TGS experiment with $E(\infty)$ the concentration of the largest particles almost doubles compared to the experiment without vdW enhancement. Table 3 lists the global and annual mean values of relevant quantities, revealing that the shift towards larger particle size by the vdW enhancement results in a reduction of the stratospheric aerosol burden, the AOD at 550 nm, and a significant change in the radiative forcing. Since the SW forcing decreases more strongly than the LW forcing (in absolute values), the net radiative forcing becomes considerably less negative and can even become positive in the 50_TGS experiment with $E(\infty)$. We may therefore conclude that the strong decrease in forcing efficiency for larger injection rates observed in the experiments without vdW enhancement is probably underestimated if we were to account fully for the effect of vdW forces on coagulation. But we should note that the exact impact of vdW forces on coagulation is not very well determined from theory and laboratory experiments and that the implementation in our model is only tentative.

3.6 Effect of radiatively interactive aerosol

We now discuss the impact of having radiatively interactive aerosols in our model simulations. Indeed the heating of the stratosphere and the upper troposphere described above can be expected to have a considerable impact on the atmospheric dynamics and thereby on the distribution and evolution of the aerosol distribution itself. In order to quantify this impact, we performed an SAI simulation called NORAD in which the aerosol does not interact with radiation (only the instantaneous radiative forcing is computed from a double radiation call but the model integration is performed with stratospheric aerosols that are invisible to the radiation). In this scenario, we chose a relatively high injection height of 21 km, because without any aerosol-induced heating an important factor for the vertical transport from the tropical tropopause layer (TTL) to the stratosphere is missing.

The resulting vertical distributions of the aerosol extinction coefficient with and without radiative interaction are shown in Fig. 14. The heating causes the aerosol to rise higher and spread more meridionally, as it was already described by Aquila et al. (2012) for model simulations of the 1991 Mount Pinatubo eruption. This may also be related to the changes in stratospheric dynamics described below. The spatial distribution in the NORAD (at 21 km) experiment is closer to the one in the 17_KM or 19_KM experiments, but with a more pronounced maximum in extinction coefficient. Due to the more confined spatial distribution in the NORAD simulation, local concentrations of sulfuric acid and particles are larger, causing the particles to grow larger through condensation and coagulation. Many of the larger particles leave the stratosphere through sedimentation already in the tropics, so that the stratospheric aerosol burden in the NORAD experiment is smaller than in the 21_KM, 19_KM and even 17_KM experiments. Therefore, the resulting global mean net radiative forcing is significantly smaller in the NORAD experiment (–0.9 $\mathrm{W\,m^{-2}}$) than in the 21_KM simulation with radiatively interactive aerosols (–1.4 $\mathrm{W\,m^{-2}}$).

3.7 Impact on the QBO

The locally very strong heating of the lower stratosphere and upper troposphere due to the aerosol-radiation interactions (up to 16 K for the standard SAI scenario relative to the control run) can be expected to have a considerable impact on atmospheric dynamics. The quasi-biennial oscillation (QBO) in the equatorial stratosphere is an important dynamical feature of the stratosphere. It consists of easterly and westerly winds alternating with a period of approximately 28 months. The phase of the QBO is known to affect the poleward transport of trace gases and aerosols in the stratosphere (Trepte and Hitchman, 1992).
Indeed Fig. 15 shows that the QBO is strongly affected by the sulfur aerosol injection, in agreement with a previous study by Aquila et al. (2014). While the QBO period in the spin up and CONTROL simulations including only stratospheric background aerosol is approximately 28 months (varying between 24 and 32 months as the background aerosol layer builds up), it increases significantly already for the smallest simulated injection rate of 2 $\mathrm{Tg\,S\,yr^{-1}}$. In the 5 $\mathrm{Tg\,S\,yr^{-1}}$ scenario the periodicity disappears, while easterly winds dominate below 25 km and westerly winds dominate above. For the scenarios with 10 $\mathrm{Tg\,S\,yr^{-1}}$ and more, the direction of the propagation inverts from downward to upward, but with a period of approximately one year and with smaller amplitude.

3.8 Sulfate impact at the Earth's surface

The deposition of acids at the land or ocean surface can be harmful to ecosystems. Although the additional input of sulfur species to the Earth system is (at least for the lower emission scenarios) not large compared to the already existing anthropogenic and natural inputs (of about 136 $\mathrm{Tg\,S\,yr^{-1}}$ (Kravitz et al., 2009)) sulfate SAI constitutes an additional and intentional sulfur input. Therefore, it is important to study the deposition fluxes of sulfuric acid due to SAI. Before their deposition sulfate particles near the surface can also have a negative impact on human health by increasing particle concentrations in ambient air ($\mathrm{PM_{2.5}}$).

Due to the relatively short lifetime of the sulfate particles in the troposphere, the relevance and relative importance of the various processes at play are different than in the stratosphere. Therefore, we decided not to activate the microphysical processes of coagulation, nucleation, condensation and evaporation below the tropopause. Thus, the particles do not grow or shrink any

more, but they are removed via wet and dry deposition (both independent of the particle size) and sedimentation.
However, our model setup makes it possible to study in first approximation the contribution of SAI to increase particle concentration at the Earth's surface (i.e., in the lowest atmospheric model layer) as well as the deposition rate of aerosols to the ground or sea surface.

The deposition rate of sulfate aerosol coming from the stratosphere to the surface in the STANDARD scenario is shown in Fig. 16. It is largest in the low and mid-latitudes and over the ocean, where it reaches up to $82\,\mathrm{mg\,S\,m^{-2}\,yr^{-1}}$. This spatial distribution is similar to the one shown by Kravitz et al. (2009). However, the absolute values of the deposition anomaly shown by Kravitz et al. (2009) are larger than in our simulations because their anomaly includes changes in the deposition of tropospheric sulfur (between two simulations with and without SAI), which is not the case in our model. On a global average, wet deposition contributes 68 %, dry deposition 28 %, and sedimentation 4 % to the total deposition rate at the surface.
For estimating the impact in terms of acidic deposition on ecosystems, we convert the deposition rate of sulfur from $\mathrm{mg\,S\,m^{-2}\,yr^{-1}}$ to $\mathrm{mEq\,m^{-2}\,yr^{-1}}$ (used in critical loading studies like Kuylenstierna et al. (2001), cited by Kravitz et al. (2009)) using a conversion factor of 16 mg S = 1 mEq. With this, the largest additional deposition rates (due to aerosol from SAI) in our simulation are equivalent to $5\,\mathrm{mEq\,m^{-2}\,yr^{-1}}$. This is well below the critical loadings for almost all ecosystems reported by Kuylenstierna et al. (2001) and on the very low side for the most sensitive type of ecosystems (e.g., waterways in Sweden, which have a critical load of 1–44 $\mathrm{mEq\,m^{-2}\,yr^{-1}}$).

Kravitz et al. (2009) gives a global mean value of $270\,\mathrm{mg\,S\,m^{-2}\,yr^{-1}}$ for the present-day deposition rate of tropospheric sulfur. Compared with this large number, our simulated global mean additional sulfate deposition due to SAI of $18\,\mathrm{mg\,S\,m^{-2}\,yr^{-1}}$, but even the maximum value of $82\,\mathrm{mg\,S\,m^{-2}\,yr^{-1}}$ is rather small.
In order to assess the possible impact on human health, we also compute the concentration of $PM_{2.5}$ at ground level due to aerosol particles coming from the stratosphere. For this we take the sum over the aerosol mass in all size bins with dry diameter lower than 2.5 µm, assuming that they consist of ammonium sulfate with a density of $1.77\,\mathrm{g\,cm^{-3}}$. Due to this choice, we probably overestimate $PM_{2.5}$, because the wet radius of some of these particles is actually above 2.5 µm. Allowing the particles to grow further in the troposphere would also result in smaller concentrations of small particles.
The additional $PM_{2.5}$ concentration at the surface due to SAI is shown in Fig. 17. It does not exceed $1\,\mathrm{\mu g\,m^{-3}}$ and is largest over the ocean, where the model assumes the lowest dry deposition velocities, thereby increasing the aerosol lifetime in marine boundary layer. In order to get a rough estimation of the impact of this additional $PM_{2.5}$ on human health, we first compute the product of $PM_{2.5}$ from SAI and human population count (2015 values from SSP1, see Riahi et al. (2017)) at the grid cell level and cumulate these values over the whole Earth. This cumulative sum is $4.2 \cdot 10^9\,\mathrm{\mu g\,m^{-3}\,person}$, which is approximately 2% of the value found using present day $PM_{2.5}$ values from van Donkelaar et al. (2016) at $2.4 \cdot 10^{11}\,\mathrm{\mu g\,m^{-3}\,person}$. Although previous studies (e.g. Partanen et al., 2013) have translated such changes in surface PM concentrations into mortality or morbidity rates, we do not attempt to do so as the health impact of PM is dependent on aerosol size and chemical composition in ways that are unknown.

4 Conclusions

The model results for various scenarios of stratospheric aerosol injections (SAI) presented in this study imply that the net radiative forcing achievable through equatorial SAI with SO_2 might be smaller than previously estimated. The radiative heating through the aerosol can disturb the stratospheric dynamics in such a way that the meridional transport is hindered, resulting in larger sulfate concentrations in the tropics, which enhances particle growth. The larger particles are responsible for an important positive LW forcing, which can compensate the negative SW forcing (cooling) almost completely for large SO_2 injection rates like $50\,\mathrm{Tg\,S\,yr^{-1}}$ (the maximum rate simulated in this study). We find that it might be impossible to achieve a more negative net instantaneous radiative forcing than –2 $\mathrm{W\,m^{-2}}$ with equatorial SO_2 injections.

We also find that SO_2 injections at higher altitude (in the range 17 to 23 km) do not result in larger (i.e., more negative) radiative forcing, because the particles grow to larger size during their stratospheric lifetime and have less optimal optical properties. This finding contradicts previous studies by ~~English et al. (2013)~~ Niemeier et al. (2011) and Niemeier and Timmreck (2015) but agrees with a recent study by Niemeier and Schmidt (2017).

Enlarging the injection area from one equatorial grid cell to several grid cells between 30°S and 30°N resulted in smaller particles, but also in a smaller global aerosol burden, which in total causes the net radiative forcing to be equal to the one from equatorial injections. Restricting the injections to a shorter period of the year with seasonally varying latitude resulted in a small increase in net radiative forcing, but probably only due to stronger updraft (more heating from larger sulfate concentrations).

The simulated impact on stratospheric dynamics through radiative heating by the aerosol agrees with a previous study by Aquila et al. (2014) in that the QBO breaks down for injection rates larger than about $5\,\mathrm{Tg\,S\,yr^{-1}}$. This results in a stronger subtropical transport barrier, which causes the particles to grow even larger. In contrast, the impact of the additional sulfate at the Earth's surface is probably small. The maximum additional acidic deposition is below the critical loadings for almost all ecosystems and the additional particle concentrations ($PM_{2.5}$) are below $1\,\mathrm{\mu g\,m^{-3}}$, resulting in a small 2 % increase of population-weighted $PM_{2.5}$ relative to present-day conditions.

In this study we only consider the instantaneous radiative forcing (IRF) by the aerosol. But in the real climate system this IRF will induce rapid adjustments and slower feedback mechanisms. The combination of these ultimately determines the impact of SAI on the climate. For studying the slow feedbacks one would need to couple the atmospheric model at least to an ocean model, which we did not do in the presented experiments. But we studied the rapid adjustments to the IRF, resulting in an effective radiative forcing (ERF), in a separate paper (Boucher et al., 2017). We found that rapid adjustments significantly enhance ERF relative to IRF because of stratospheric warming and despite a moistening of the stratosphere.

In our experiments we found that the positive LW forcing can compensate a large part of the negative SW forcing, i.e. 67 % in the STANDARD $10\,\mathrm{Tg\,S\,yr^{-1}}$ and 93 % in the $50\,\mathrm{Tg\,S\,yr^{-1}}$ scenario. Therefore, we argue that the LW forcing should always be considered in modelling studies of SAI, which has not often been the case in the past. Considering only the SW forcing of SAI will lead to considerable overestimation of its efficacy.

~~As the~~ The simulation results depend largely on the modelled evolution of particle size, radiative heating and stratospheric dynamics (controlling the spatial distribution). The stratospheric processes determining this are complex and uncertain and

their implementation causes considerable disagreement between different models. In contrast to volcanic eruptions, models cannot be constrained by observations in the case of purely hypothetical geoengineering scenarios. Therefore, it would be worthwhile to compare ~~them with~~ the results with those from other models, like we did it in Sec. 3.2. Further intercomparison could increase the robustness of the findings from this study and it could also help to further improve the models by examining differences (e.g. the refractive index and the resulting optical properties of the aerosol) in more detail.

In additional sensitivity experiments including the effect of van der Waals (vdW) forces on coagulation we found that the strong decrease in forcing efficiency for larger injection rates observed in the other experiments (without vdW enhancement) is probably underestimated. But we should note that the exact impact of vdW forces on coagulation is not very well determined from theory and laboratory experiments and that the implementation in our model is only tentative. These results thus justify further work on parametrising aerosol coagulation in models as well as our choice for a sectional approach to represent the aerosol size dynamics.

It has to be noted that the results presented in the previous section are strongly influenced by the choice of the injection scenarios. Spreading the injections over larger areas would lower the local concentrations of precursor gases and sulfate and would therefore probably result in less particle growth and more favourable optical properties. Smaller local extinction by the aerosol would also cause less heating and probably less updraft of the particles, which may increase the importance of the initial injection height. Hence, our finding that higher injections do not result in larger radiative forcing (for a given sulfur mass), may not hold for different injection schemes.

We also have to keep in mind that our model setup does not include chemical feedbacks of the aerosol, e.g. on stratospheric ozone. Induced changes in ozone concentration might alter the radiative effects of SAI (especially the stratospheric heating) considerably. The prescription of sea surface temperatures constitutes another limitation for correctly simulating the feedback of SAI on atmospheric dynamics.

The phenomenon of aerosol lifting through local heating and dynamical changes might also be used for the design of better injection strategies, as it could allow technically less demanding injections at lower altitudes. But as the larger local sulfate concentrations needed for the updraft will probably also enhance particle growth, it is unclear whether such a strategy could be more efficient at all. One could possibly imagine methods to increase buoyancy in the initial aerosol (or aerosol precursor) plume that have less impact on particle growth.

Overall, this study may provide additional evidence that solar radiation management with stratospheric sulfate aerosols (formed from tropical injections of SO_2) is still more complicated, probably less effective and may implicate stronger side-effects than initially thought.

Code and data availability. The data are available from the authors upon request. The code of S3A can be downloaded along with the LMDZ model from http://lmdz.lmd.jussieu.fr. S3A code is mostly contained within a separate directory StratAer of the model physics and is activated at compilation with a CPP key. A model configuration LMDZORSTRATAER_v6 containing the S3A module is also available within the modipsl/libIGCM model environment of the IPSL Earth System Model http://forge.ipsl.jussieu.fr/igcmg_doc.

Author contributions. C. Kleinschmitt developed most of the S3A model, designed and performed the SAI simulations, and analysed the data. He wrote most of the article. O. Boucher and U. Platt assisted in the model development and the analysis of the model simulations.

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References

Aquila, V., Oman, L. D., Stolarski, R. S., Colarco, P. R., and Newman, P. A.: Dispersion of the volcanic sulfate cloud from a Mount Pinatubo–like eruption, Journal of Geophysical Research: Atmospheres, 117, D06 216, doi:10.1029/2011JD016968, 2012.

Aquila, V., Garfinkel, C. I., Newman, P., Oman, L., and Waugh, D.: Modifications of the quasi-biennial oscillation by a geoengineering perturbation of the stratospheric aerosol layer, Geophysical Research Letters, 41, 1738–1744, doi:10.1002/2013GL058818, 2014.

Bekki, S.: Oxidation of volcanic SO_2: A sink for stratospheric OH and H_2O, Geophysical Research Letters, 22, 913–916, doi:10.1029/95GL00534, 1995.

Boucher, O., Kleinschmitt, C., and Myhre, G.: Quasi-additivity of the radiative effects of marine cloud brightening and stratospheric sulfate aerosol injection, Geophysical Research Letters, 44, 2017GL074 647, doi:10.1002/2017GL074647, in press, 2017.

Budyko, M. I.: Climatic Changes, American Geophysical Union, doi:10.1029/SP010, 1977.

Chan, T. W. and Mozurkewich, M.: Measurement of the coagulation rate constant for sulfuric acid particles as a function of particle size using tandem differential mobility analysis, Journal of Aerosol Science, 32, 321–339, doi:10.1016/S0021-8502(00)00081-1, 2001.

Crutzen, P. J.: Albedo enhancement by stratospheric sulfur injections: A contribution to resolve a policy dilemma?, Climatic Change, 77, 211–219, doi:10.1007/s10584-006-9101-y, 2006.

de la Cámara, A., Lott, F., and Abalos, M.: Climatology of the middle atmosphere in LMDz: Impact of source-related parameterizations of gravity wave drag, Journal of Advances in Modeling Earth Systems, pp. 1507–1525, doi:10.1002/2016MS000753, 2016.

Dykema, J. A., Keith, D. W., and Keutsch, F. N.: Improved aerosol radiative properties as a foundation for solar geoengineering risk assessment, Geophysical Research Letters, 43, 7758–7766, doi:10.1002/2016GL069258, 2016.

English, J. M., Toon, O. B., and Mills, M. J.: Microphysical simulations of sulfur burdens from stratospheric sulfur geoengineering, Atmospheric Chemistry and Physics, 12, 4775–4793, doi:10.5194/acp-12-4775-2012, 2012.

English, J. M., Toon, O. B., and Mills, M. J.: Microphysical simulations of large volcanic eruptions: Pinatubo and Toba, Journal of Geophysical Research: Atmospheres, 118, 1880–1895, doi:10.1002/jgrd.50196, 2013.

Fahey, D. W., Kawa, S. R., Woodbridge, E. L., Tin, P., Wilson, J. C., Jonsson, H. H., Dye, J. E., Baumgardner, D., Borrmann, S., Toohey, D. W., Avallone, L. M., Proffitt, M. H., Margitan, J., Loewenstein, M., Podolske, J. R., Salawitch, R. J., Wofsy, S. C., Ko, M. K. W., Anderson, D. E., Schoeber, M. R., and Chan, K. R.: In situ measurements constraining the role of sulphate aerosols in mid-latitude ozone depletion, Nature, 363, 509–514, doi:10.1038/363509a0, 1993.

Ferraro, A. J., Highwood, E. J., and Charlton-Perez, A. J.: Stratospheric heating by potential geoengineering aerosols, Geophysical Research Letters, 38, L24 706, doi:10.1029/2011GL049761, 2011.

Heckendorn, P., Weisenstein, D., Fueglistaler, S., Luo, B. P., Rozanov, E., Schraner, M., Thomason, L. W., and Peter, T.: The impact of geoengineering aerosols on stratospheric temperature and ozone, Environmental Research Letters, 4, 045 108, doi:10.1088/1748-9326/4/4/045108, 2009.

Hourdin, F., Musat, I., Bony, S., Braconnot, P., Codron, F., Dufresne, J.-L., Fairhead, L., Filiberti, M.-A., Friedlingstein, P., Grandpeix, J.-Y., Krinner, G., LeVan, P., Li, Z.-X., and Lott, F.: The LMDZ4 general circulation model: climate performance and sensitivity to parametrized physics with emphasis on tropical convection, Climate Dynamics, 27, 787–813, doi:10.1007/s00382-006-0158-0, 2006.

Hourdin, F., Foujols, M.-A., Codron, F., Guemas, V., Dufresne, J.-L., Bony, S., Denvil, S., Guez, L., Lott, F., Ghattas, J., Braconnot, P., Marti, O., Meurdesoif, Y., and Bopp, L.: Impact of the LMDZ atmospheric grid configuration on the climate and sensitivity of the IPSL-CM5A coupled model, Climate Dynamics, 40, 2167–2192, doi:10.1007/s00382-012-1411-3, 2013.

Irvine, P. J., Kravitz, B., Lawrence, M. G., and Muri, H.: An overview of the Earth system science of solar geoengineering, Wiley Interdisciplinary Reviews: Climate Change, 7, 815–833, doi:10.1002/wcc.423, 2016.

Jacobson, M. Z., Turco, R. P., Jensen, E. J., and Toon, O. B.: Modeling coagulation among particles of different composition and size, Atmospheric Environment, 28, 1327–1338, doi:10.1016/1352-2310(94)90280-1, 1994.

Keith, D. W., Weisenstein, D. K., Dykema, J. A., and Keutsch, F. N.: Stratospheric solar geoengineering without ozone loss, Proceedings of the National Academy of Sciences, 113, 14 910–14 914, doi:10.1073/pnas.1615572113, 2016.

Kleinschmitt, C., Boucher, O., Bekki, S., Lott, F., and Platt, U.: The Sectional Stratospheric Sulfate Aerosol module (S3A-v1) within the LMDZ general circulation model: description and evaluation against stratospheric aerosol observations, Geoscientific Model Development, 10, 3359–3378, doi:10.5194/gmd-10-3359-2017, 2017.

Kravitz, B., Robock, A., Oman, L., Stenchikov, G., and Marquardt, A. B.: Sulfuric acid deposition from stratospheric geoengineering with sulfate aerosols, Journal of Geophysical Research: Atmospheres, 114, D14 109, doi:10.1029/2009JD011918, 2009.

Kravitz, B., Caldeira, K., Boucher, O., Robock, A., Rasch, P. J., Alterskjær, K., Karam, D. B., Cole, J. N. S., Curry, C. L., Haywood, J. M., Irvine, P. J., Ji, D., Jones, A., Kristjánsson, J. E., Lunt, D. J., Moore, J. C., Niemeier, U., Schmidt, H., Schulz, M., Singh, B., Tilmes, S., Watanabe, S., Yang, S., and Yoon, J.-H.: Climate model response from the Geoengineering Model Intercomparison Project (GeoMIP), Journal of Geophysical Research: Atmospheres, 118, 8320–8332, doi:10.1002/jgrd.50646, 2013.

Kuylenstierna, J. C. I., Rodhe, H., Cinderby, S., and Hicks, K.: Acidification in Developing Countries: Ecosystem Sensitivity and the Critical Load Approach on a Global Scale, Ambio, 30, 20–28, doi:10.1579/0044-7447-30.1.20, 2001.

Laakso, A., Korhonen, H., Romakkaniemi, S., and Kokkola, H.: Radiative and climate effects of stratospheric sulfur geoengineering using seasonally varying injection areas, Atmospheric Chemistry and Physics, 17, 6957–6974, doi:10.5194/acp-17-6957-2017, 2017.

MacMartin, D. G., Kravitz, B., Long, J. C. S., and Rasch, P. J.: Geoengineering with stratospheric aerosols: What do we not know after a decade of research?, Earth's Future, 4, 543–548, doi:10.1002/2016EF000418, 2016.

McClellan, J., Keith, D. W., and Apt, J.: Cost analysis of stratospheric albedo modification delivery systems, Environmental Research Letters, 7, 034 019, doi:10.1088/1748-9326/7/3/034019, 2012.

Niemeier, U. and Schmidt, H.: Changing transport processes in the stratosphere by radiative heating of sulfate aerosols, Atmospheric Chemistry and Physics Discussions, 2017, 1–24, doi:10.5194/acp-2017-470, 2017.

Niemeier, U. and Timmreck, C.: What is the limit of climate engineering by stratospheric injection of SO_2?, Atmospheric Chemistry and Physics, 15, 9129–9141, doi:10.5194/acp-15-9129-2015, 2015.

Niemeier, U., Schmidt, H., and Timmreck, C.: The dependency of geoengineered sulfate aerosol on the emission strategy, Atmospheric Science Letters, 12, 189–194, doi:10.1002/asl.304, 2011.

Partanen, A. I., Laakso, A., Schmidt, A., Kokkola, H., Kuokkanen, T., Pietikäinen, J.-P., Kerminen, V.-M., Lehtinen, K. E. J., Laakso, L., and Korhonen, H.: Climate and air quality trade-offs in altering ship fuel sulfur content, Atmospheric Chemistry and Physics, 13, 12 059–12 071, doi:10.5194/acp-13-12059-2013, 2013.

Pierce, J. R., Weisenstein, D. K., Heckendorn, P., Peter, T., and Keith, D. W.: Efficient formation of stratospheric aerosol for climate engineering by emission of condensible vapor from aircraft, Geophysical Research Letters, 37, L18 805, doi:10.1029/2010GL043975, 2010.

Pitari, G., Aquila, V., Kravitz, B., Robock, A., Watanabe, S., Cionni, I., Luca, N. D., Genova, G. D., Mancini, E., and Tilmes, S.: Stratospheric ozone response to sulfate geoengineering: Results from the Geoengineering Model Intercomparison Project (GeoMIP), Journal of Geophysical Research: Atmospheres, 119, 2629–2653, doi:10.1002/2013JD020566, 2014.

Punge, H. J., Konopka, P., Giorgetta, M. A., and Müller, R.: Effects of the quasi-biennial oscillation on low-latitude transport in the stratosphere derived from trajectory calculations, Journal of Geophysical Research: Atmospheres, 114, D03 102, doi:10.1029/2008JD010518, 2009.

Rasch, P. J., Crutzen, P. J., and Coleman, D. B.: Exploring the geoengineering of climate using stratospheric sulfate aerosols: The role of particle size, Geophysical Research Letters, 35, L02 809, doi:10.1029/2007GL032179, 2008.

Reichler, T., Dameris, M., and Sausen, R.: Determining the tropopause height from gridded data, Geophysical Research Letters, 30, 2042, doi:10.1029/2003GL018240, 2003.

Riahi, K., van Vuuren, D. P., Kriegler, E., Edmonds, J., O'Neill, B. C., Fujimori, S., Bauer, N., Calvin, K., Dellink, R., Fricko, O., Lutz, W., Popp, A., Cuaresma, J. C., KC, S., Leimbach, M., Jiang, L., Kram, T., Rao, S., Emmerling, J., Ebi, K., Hasegawa, T., Havlik, P., Humpenöder, F., Silva, L. A. D., Smith, S., Stehfest, E., Bosetti, V., Eom, J., Gernaat, D., Masui, T., Rogelj, J., Strefler, J., Drouet, L., Krey, V., Luderer, G., Harmsen, M., Takahashi, K., Baumstark, L., Doelman, J. C., Kainuma, M., Klimont, Z., Marangoni, G., Lotze-Campen, H., Obersteiner, M., Tabeau, A., and Tavoni, M.: The Shared Socioeconomic Pathways and their energy, land use, and greenhouse gas emissions implications: An overview, Global Environmental Change, 42, 153–168, doi:10.1016/j.gloenvcha.2016.05.009, 2017.

Ricke, K. L., Morgan, M. G., and Allen, M. R.: Regional climate response to solar-radiation management, Nature Geoscience, 3, 537–541, doi:10.1038/ngeo915, 2010.

Robock, A., Oman, L., and Stenchikov, G. L.: Regional climate responses to geoengineering with tropical and Arctic SO_2 injections, Journal of Geophysical Research: Atmospheres, 113, D16 101, doi:10.1029/2008JD010050, 2008.

Santer, B. D., Wehner, M. F., Wigley, T. M. L., Sausen, R., Meehl, G. A., Taylor, K. E., Ammann, C., Arblaster, J., Washington, W. M., Boyle, J. S., and Brüggemann, W.: Contributions of Anthropogenic and Natural Forcing to Recent Tropopause Height Changes, Science, 301, 479–483, doi:10.1126/science.1084123, 2003.

Schulz, M., Textor, C., Kinne, S., Balkanski, Y., Bauer, S., Berntsen, T., Berglen, T., Boucher, O., Dentener, F., Guibert, S., Isaksen, I. S. A., Iversen, T., Koch, D., Kirkevåg, A., Liu, X., Montanaro, V., Myhre, G., Penner, J. E., Pitari, G., Reddy, S., Seland, Ø., Stier, P., and Takemura, T.: Radiative forcing by aerosols as derived from the AeroCom present-day and pre-industrial simulations, Atmospheric Chemistry and Physics, 6, 5225–5246, doi:10.5194/acp-6-5225-2006, 2006.

Sekiya, T., Sudo, K., and Nagai, T.: Evolution of stratospheric sulfate aerosol from the 1991 Pinatubo eruption: Roles of aerosol microphysical processes, Journal of Geophysical Research: Atmospheres, 121, 2911–2938, doi:10.1002/2015JD024313, 2016.

Tilmes, S., Müller, R., and Salawitch, R.: The Sensitivity of Polar Ozone Depletion to Proposed Geoengineering Schemes, Science, 320, 1201–1204, doi:10.1126/science.1153966, 2008.

Tilmes, S., Garcia, R. R., Kinnison, D. E., Gettelman, A., and Rasch, P. J.: Impact of geoengineered aerosols on the troposphere and stratosphere, Journal of Geophysical Research: Atmospheres, 114, D12 305, doi:10.1029/2008JD011420, 2009.

Trepte, C. R. and Hitchman, M. H.: Tropical stratospheric circulation deduced from satellite aerosol data, Nature, 355, 626–628, doi:10.1038/355626a0, 1992.

van Donkelaar, A., Martin, R. V., Brauer, M., Hsu, N. C., Kahn, R. A., Levy, R. C., Lyapustin, A., Sayer, A. M., and Winker, D. M.: Global Estimates of Fine Particulate Matter using a Combined Geophysical-Statistical Method with Information from Satellites, Models, and Monitors, Environmental Science & Technology, 50, 3762–3772, doi:10.1021/acs.est.5b05833, 2016.

Visioni, D., Pitari, G., and Aquila, V.: Sulfate geoengineering: a review of the factors controlling the needed injection of sulfur dioxide, Atmospheric Chemistry and Physics, 17, 3879–3889, doi:10.5194/acp-17-3879-2017, 2017.

Weisenstein, D. K., Keith, D. W., and Dykema, J. A.: Solar geoengineering using solid aerosol in the stratosphere, Atmospheric Chemistry and Physics, 15, 11 835–11 859, doi:10.5194/acp-15-11835-2015, 2015.

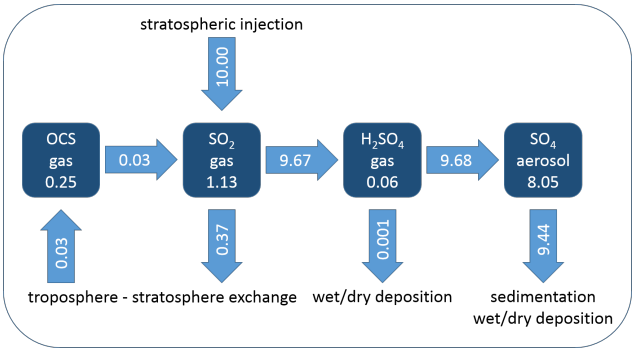

Figure 1. Annual mean stratospheric burdens (boxes, in $\mathrm{Tg\,S}$) and fluxes (arrows, in $\mathrm{Tg\,S\,yr^{-1}}$) of the represented sulfur species from the 5$^{\mathrm{th}}$ year (quasi-steady state) of the STANDARD experiment. Minor discrepancies in the fluxes indicate that the steady state is not entirely reached after 5 years.

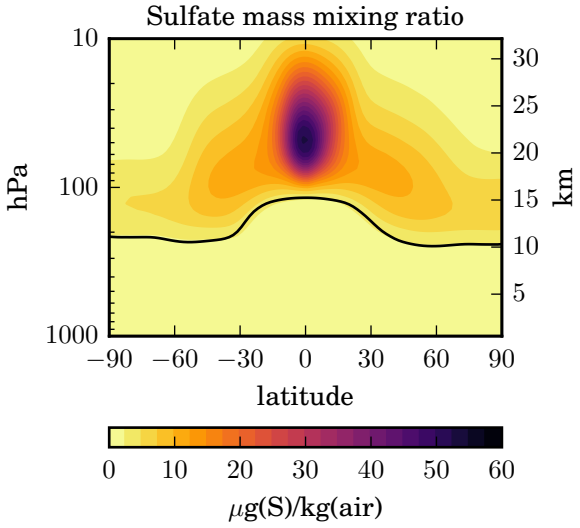

Figure 2. Latitude-height cross-section of the zonal and annual mean sulfate aerosol mass mixing ratio (in $\mu g\,S\,kg\,air^{-1}$) for the STANDARD experiment.

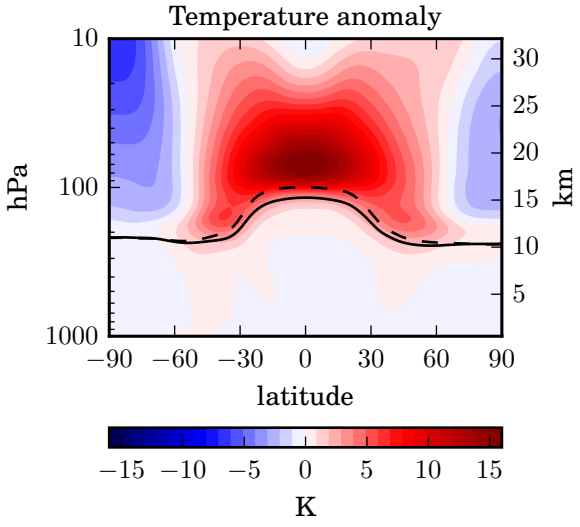

Figure 3. Latitude-height cross-section of the zonal and annual mean temperature anomaly (in K) in the STANDARD experiment relative to the CONTROL experiment.

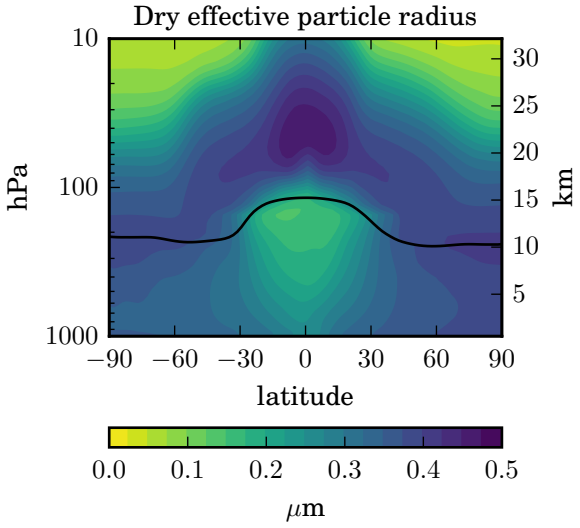

Figure 4. Latitude-height cross-section of the zonal and annual mean effective radius (in μm) of dry sulfate particles in the STANDARD experiment. The effective radius is computed from the zonal and annual means of the aerosol volume and surface area.

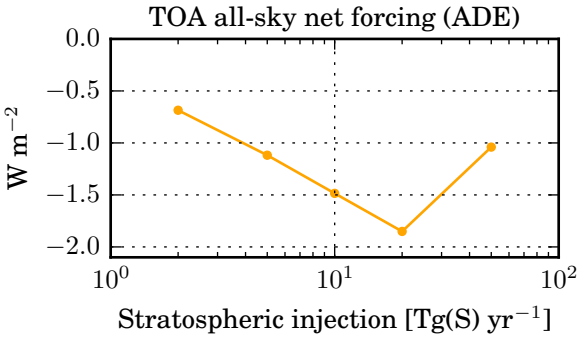

Figure 5. Global mean instantaneous net radiative forcing (aerosol direct effect = ADE) at the top of the atmosphere (TOA) in $\mathrm{W\,m^{-2}}$ for the x_TGS experiments with different injection rates.

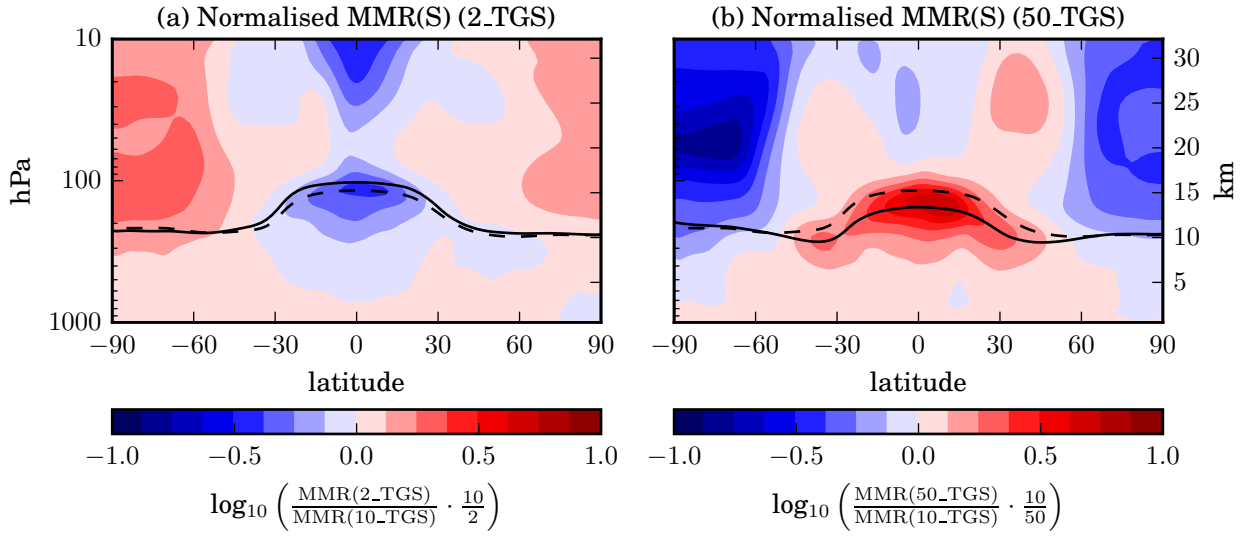

Figure 6. Latitude-height cross-sections of the zonal and annual mean sulfate aerosol mass mixing ratio (a) for the 2_TGS and (b) for the 50_TGS experiment, normalised by the value in the STANDARD (= 10_TGS) experiment and the ratio of the injection rates. The base-10 logarithm of the ratio is plotted as indicated below the color scale, so that positive (negative) values indicate larger (smaller) values than implied by a linear scaling. The mean tropopause level in the experiment is indicated by a solid line and the tropopause in the STANDARD simulation by a dashed line.

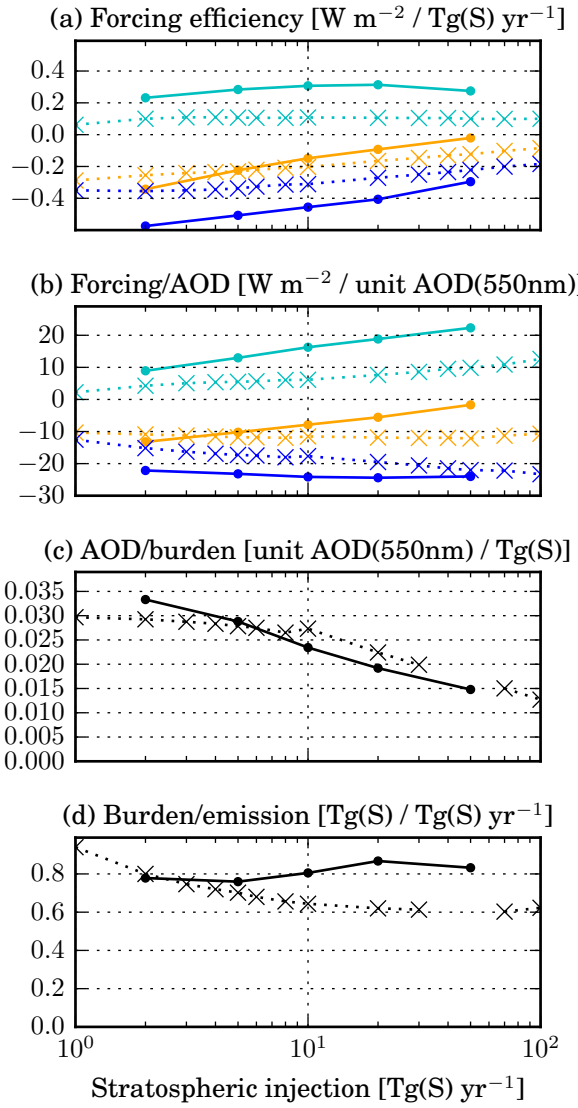

Figure 7. (a) Forcing efficiency (i.e., ratio of global mean instantaneous radiative forcing by the global sulfur injection rate) for the x_TGS simulations with different injection rates. The SW component is shown in dark blue, the LW component in light blue and the net flux in orange. Panels (b) to (d) show the decomposition of the forcing efficiency as described in the text (see Eq. 1). The solid lines show the results from the described LMDZ-S3A simulations, while the crosses show results from a previous study by Niemeier and Timmreck (2015).

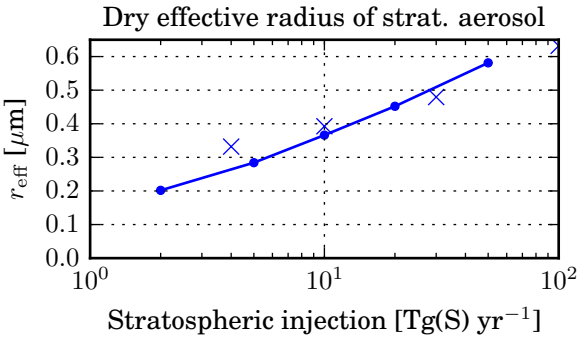

Figure 8. Global mean dry effective radius (in μm) of the stratospheric sulfate aerosol as a function of sulfur injection rate (Tg S yr^{-1}). The solid lines show the results from the x_TGS experiments using LMDZ-S3A, while the crosses show results from a previous study by Niemeier and Timmreck (2015).

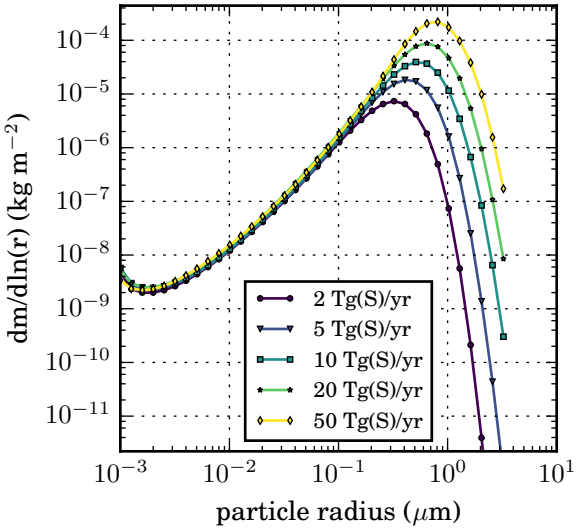

Figure 9. Column-integrated global and annual mean stratospheric aerosol mass size distribution ($\mathrm{d}m/\mathrm{d}\ln r$, in $\mathrm{kg}(\mathrm{H_2SO_4})\,\mathrm{m}^{-2}$) for the x_TGS experiments with varying sulfur injection rate.

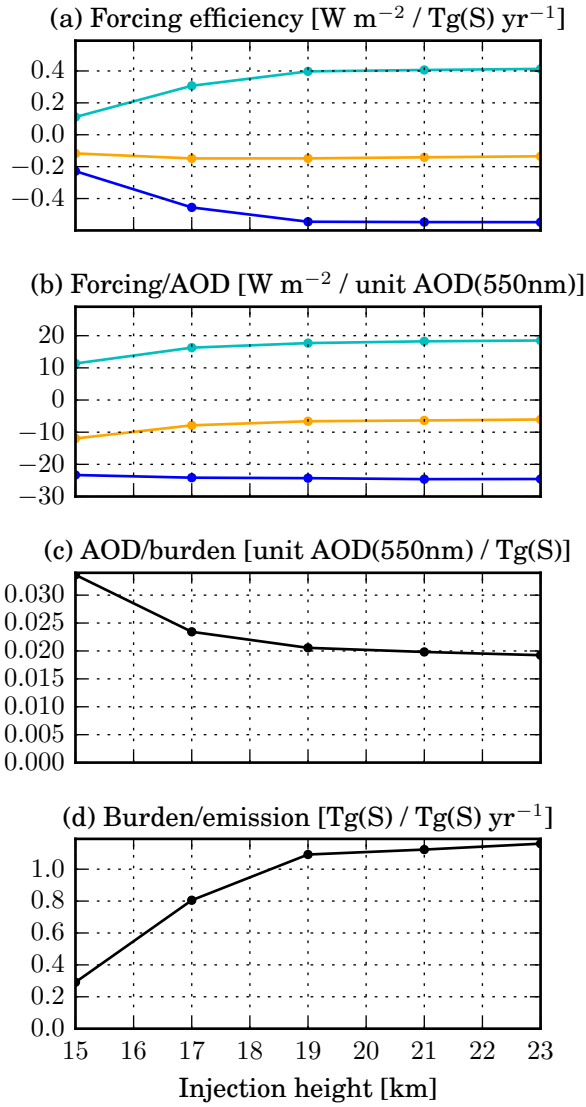

Figure 10. Forcing efficiency and its decomposition (as in Fig. 7) for the z_KM simulations with different injection heights.

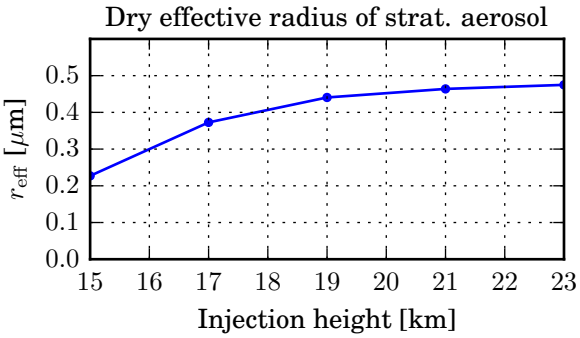

Figure 11. Global mean effective radius (μm) of the dry stratospheric sulfate aerosol as a function of injection height (in km) of the z_KM experiments.

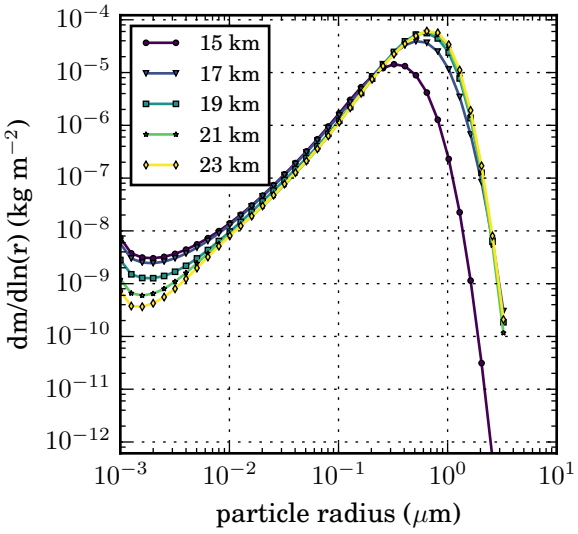

Figure 12. Column-integrated global and annual mean stratospheric aerosol mass size distribution ($\mathrm{d}m/\mathrm{d}\ln r$, in $\mathrm{kg\,m^{-2}}$) for the z_KM experiments with varying injection height.

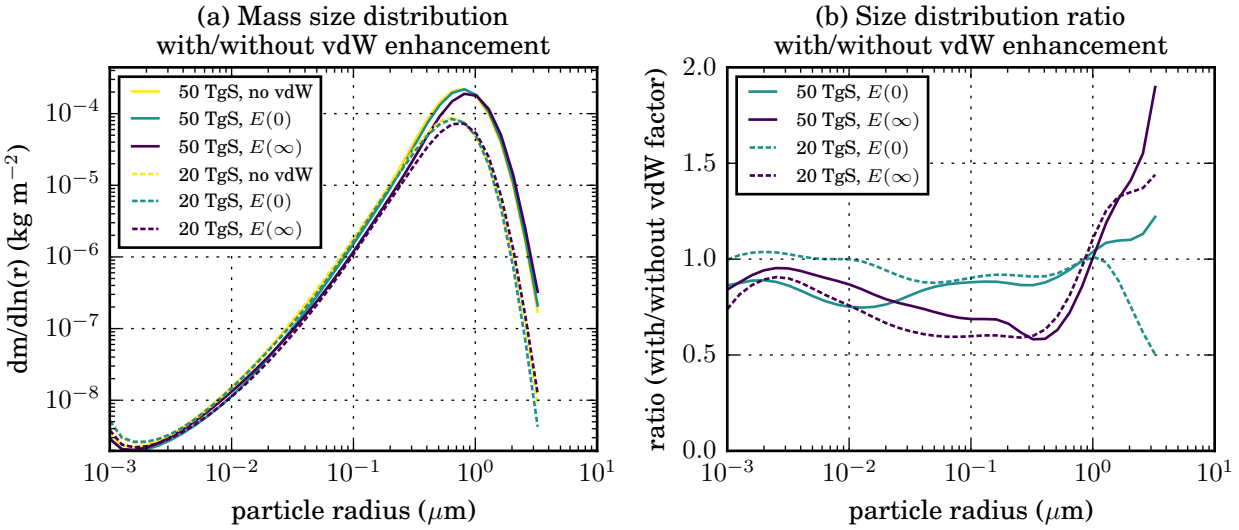

Figure 13. (a) Column-integrated global and annual mean stratospheric aerosol mass size distribution ($\mathrm{d}m/\mathrm{d}\ln r$, in $\mathrm{kg\,m^{-2}}$) for the experiments with and without van der Waals (vdW) enhancement of coagulation with the continuum regime enhancement factor $E(0)$ and the kinetic regime enhancement factor $E(\infty)$, respectively. (b) Ratio between the size distributions in the experiments with and without the vdW enhancement factor.

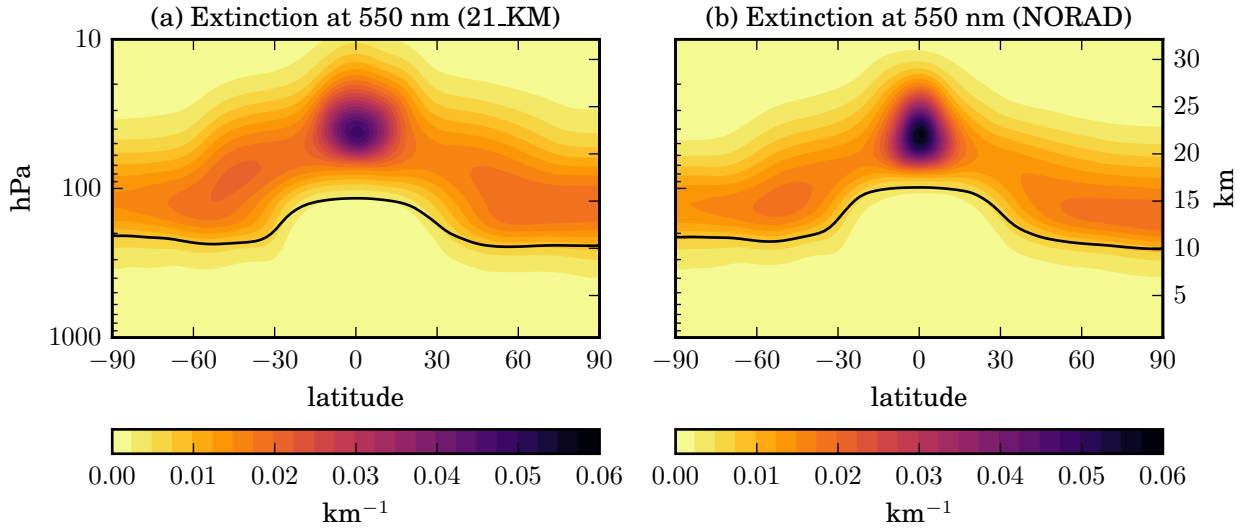

Figure 14. Latitude-height cross-section of the zonal and annual mean distribution of the aerosol extinction coefficient (km^{-1}) at 550 nm for equatorial stratospheric aerosol injections at 21 km from (a) a simulation with radiatively interactive aerosol (21_KM) and (b) a simulation with non-radiatively interactive aerosol (NORAD).

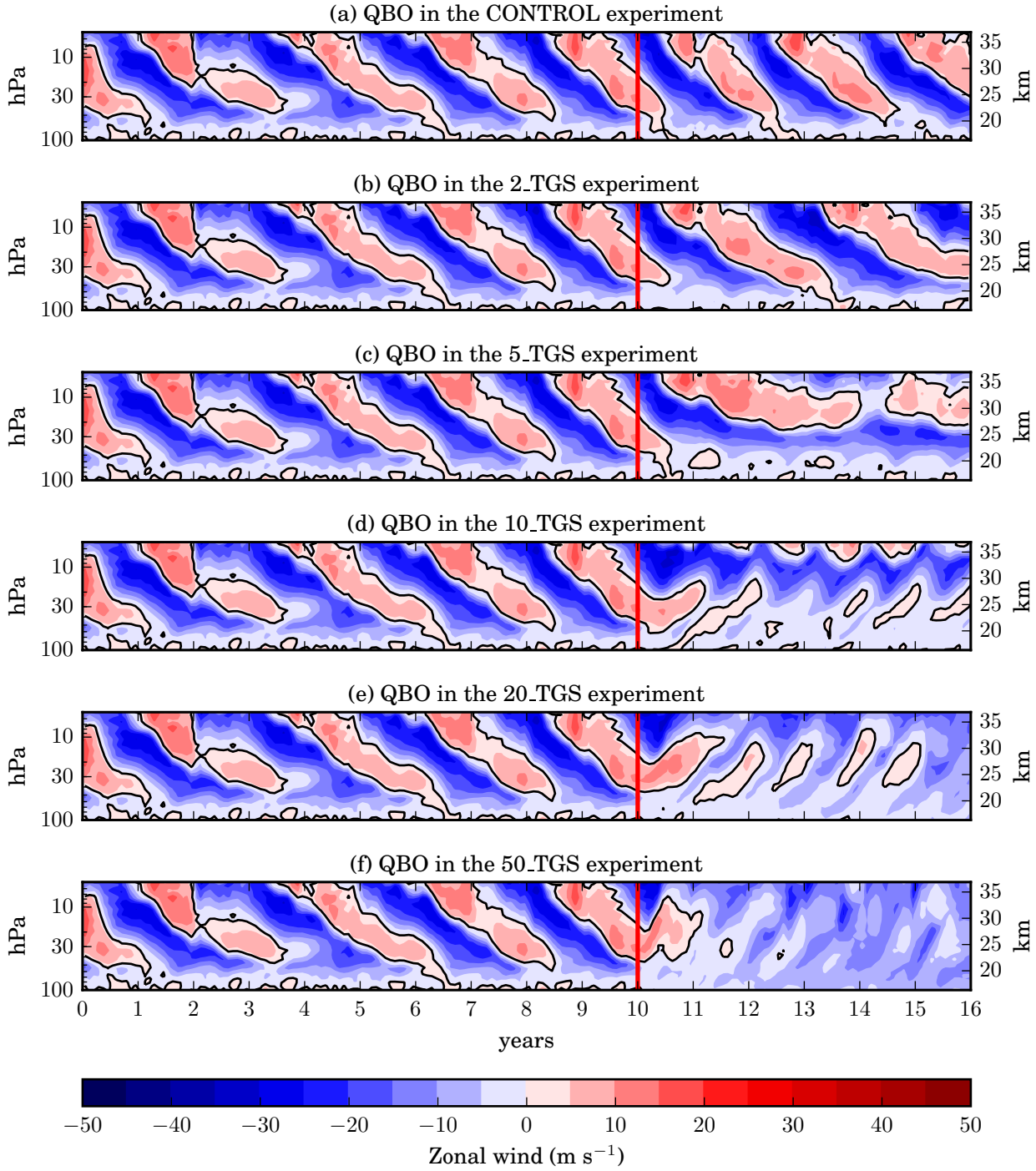

Figure 15. Temporal evolution of the vertical profiles of zonal wind, averaged zonally and between 2° N and 2° S. The QBO can be seen in the alternating downward propagation of easterly (blue) and westerly (red) wind direction. All the simulations share the same background aerosol spin up period of 10 years. The onset of the continuous sulfur injections is marked by a red line.

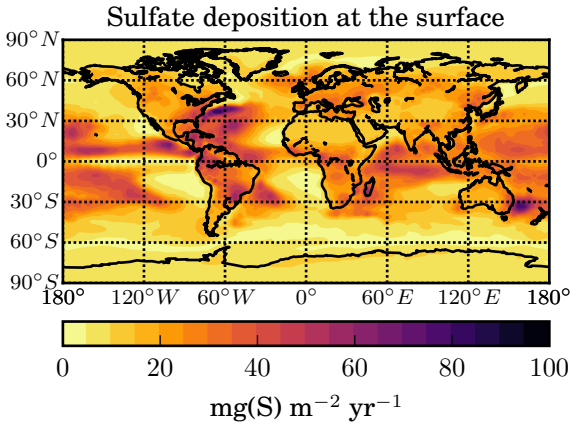

Figure 16. Annual mean deposition rate of sulfate aerosol of stratospheric origin at the Earth's surface (in $\mathrm{mg\,S\,m^{-2}\,yr^{-1}}$) in the STANDARD scenario.

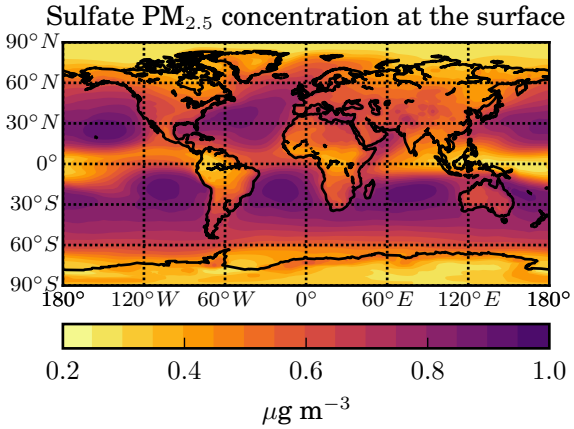

Figure 17. Annual mean concentration of sulfate aerosol particles (as ammonium sulfate) of stratospheric origin with dry diameter below 2.5 µm at the Earth's surface (in µg m^{-3}) in the STANDARD scenario.