

Answers to Reviewer 2 on the ACPD paper (acp-2017-470)

Changing transport processes in the stratosphere by radiative heating of sulfate aerosols

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We thank the reviewer for the helpful comments. We try to clarify the interpretation of the results. We include now a figure of the effective radius and move the particle number density to supplementary material. Within the supplementary material we additionally show zonal wind, residual vertical velocity, and temperature of the $10 \text{ Tg(S)} \text{y}^{-1}$ injection cases as asked by the reviewer. Accordingly, we change the discussion in the text. To strengthen our argumentation on the transport into the extratropics we add the ratio of sulfate burden in the tropics and extratropics. The ratio increases with injection rate mainly in the 60 hPa injection case, less in the 30 hPa injection case, indicating increased confinement in the 60 hPa injection.

We also add citations to previous studies which compare the impact of the vertical resolution on BDC, age of air etc. for different vertical resolutions of the mode. We say in the text that the differences between the different model resolutions can be related to the QBO but also to different vertical numerical diffusion.

Citations of the text are written in *italic* and changed or new text is highlighted in **blue**.

1. The methodology of compositing QBO phases, beginning of Section 5.1 is not clear. Transport varies with QBO phase as well as with the month of the year. The discussion in lines 260 – 270 implies that the QBO phase distinction is made for each month (correct), however it seems that in Figure 4, all months with QBOW and all months with QBOE are composited together. The caption does not say anything about which months are used in the composite, so I'm assuming that all months are used. If this is indeed the case, the plot does not show differences between QBOE and QBOW, but it shows those differences as well as differences between compositing different months of the year that coincide with QBOE and QBOE, and therefore does not answer the question that was posed. If the authors are indeed looking at the month of January or July in this Figure (the only way in which the differences between QBOE and QBOW phases can be separated clearly), then this needs to be clarified.

We add additional explanation to the definition of the composites (see also Answers to reviewer 1). We calculate the composites separately for each month but plot mostly annual mean values for zonal averages and Hovmöller diagrams for the sulfate burden. We follow the reviewers advise and plot the months of January and July instead of the annual mean in Figure 4. Additionally, we split the plot into one for the sulfate concentration and one for the vertical velocity. Overall, the result stays the same.

The description of the different transport in both composites change to: *In the tropics the meridional distribution of sulfate mass mixing ratio is broader for Comp West with less vertical extension than in Comp East (vertical cross section in Fig. 4, for January (top) and July (bottom)), illustrated also by negative anomalies above 50 hPa in the difference plots (Fig. 4, right column). The differences show higher mass mixing ratios in mid and high latitudes in Comp West which indicates stronger meridional transport in the lower stratosphere. The reason is twofold: wave propagation and different vertical velocity. Following Haynes and Shuckburgh (2000), within westerly QBO winds waves are able to propagate across the equator and break, causing mixing, on the summer side of the westerlies. This results in more meridional mixing across the subtropical transport barrier into the summer hemisphere, indicated by higher mass mixing ratios in Comp West in the summerly northern*

hemisphere in July.

Splitting the plots into January and July we now can show the different behaviour of the vertical velocity in the winter hemisphere under CE. We add for the vertical velocity: **Both months show a decrease of the downward residual vertical velocity (ω^*) poleward of 60° in the winter hemisphere, the main downward branch of the BDC.**

2. The authors in section 6 compare a simulation with 10T(s)/yr carried out with a 90-level (with QBO) and 39-level (without QBO) versions of the model in order to further demonstrate effects of the QBO. However, at no point in the manuscript do the authors show or discuss that properties of the models critical to aerosol transport are the same between the 39 and 90-level versions (except the QBO). For example, is the mean Brewer-Dobson circulation the same between these models? Is the dissipation from planetary waves (that directly impacts mixing) the same? What is the strength of stratospheric DJF and JJA jets? Are the temperature anomalies due to injections the same in both models? Without these aspects of the model demonstrated to be the same (or very similar), it is not clear here whether we're looking at differences due to the QBO or differences due to other model differences. I suggest that similarities between the aspects of the models mentioned above are shown in the appendix.

Thank you for mentioning this important point. We should have taken more care regarding this. The two versions of the model have been compared for mainly meteorological variables in Schmidt et al (2013) and related to transport by Bunzel et al (2013). Many of the requested plots can be studied in these publications. Thus, we have not added them to the appendix. Bunzel et al (2013) came to the following conclusion:

'Results of 50-yr sensitivity simulations for different climate states, performed with the state-of-the-art GCM ECHAM6 in different model configurations, have shown several similarities in the appearance of the BDC among the applied configurations of the model. The BDC pattern is qualitatively similar, independent of vertical resolution and vertical extent of the model. Even the relative contribution of resolved and unresolved waves to the driving of the total upward mass flux from the troposphere to the stratosphere is comparable. Increasing the vertical resolution in the high-top model, on the other hand, results in a slightly slower BDC. The total upward mass flux at 70 hPa is reduced by roughly 5%, and the mean age of stratospheric air increases by about 20% in the midlatitudes. We attribute the origin of this difference in age of air among the two high-top models primarily to the reduced numerical diffusion through the tropopause in the L95 model configuration.'

We added to the model description (Sect 3.1):

Bunzel and Schmidt (2013) compared simulations with low and high vertical resolution (47 and 95 levels) version of ECHAM6. The Brewer-Dobson Circulation is qualitatively similar and independent of resolution. The high resolution shows 5% less vertical mass flux and 20% increase in age of air at mid-latitudes. Numerical diffusion is reduced when increasing the vertical resolution, resulting in lower vertical extent of the sulfate layer. Schmidt et al. (2013) show that differences between the atmospheric mean states and trends simulated with two different vertical resolutions of ECHAM are in general small except for the tropics where the QBO is a dominant feature. Thus, we presume that we can assign differences between responses to sulfate aerosol forcing simulated with different vertical resolutions mostly to the internally generated or not generated QBO and to different strength of vertical numerical diffusion.

However, we cannot clearly differ between changes due the QBO or due to other impact of different vertical resolution in our results. We say in the beginning of Section 6: *Niemeier and Timmreck (2015) used ECHAM5-HAM in a 39-layer version that does not allow for an internally generated QBO, simulating instead constant equatorial easterly winds. This allows us to estimate an error in*

the sulfate forcing made by using a dynamically too simple model with, additionally, stronger numerical diffusion in vertical direction which presents itself like an additional artificial up- and downdraft. However, through the distinction of QBO phases in Section 5 we can clearly attribute effects simulated in this paper to changes in the tropical circulation. We also change the text later in Section 6, as described under 3) in this answer to reviewer.

3. Interpretation of differences between simulation 10Tg60 and Geo10 I and 10Tg30 (Figures 7 and 8) is not clear and very confusing. There is no convincing explanation in the text to account for the differences in AOD between the 3 simulations shown in Figure 7. There is an overall increase in aerosol number density in the accumulation moden Geo10 as compared to 10Tg60 (Figure 8), however if the contour level is adjusted to show the maximum contour, it could be that the distribution with latitude of aerosols looks very similar to that as in 10Tg60, and the overall change is due to something other than QBO winds.

We decide to now show the effective radius instead of the particle density, which is shifted to the supplementary material. The effective radius shows clearly an increase of the particle size. Particle size impacts the AOD, which is smaller with the same burden but larger particles.

We changed the text to: *A clear effect is the roughly 20% lower AOD in 10Tg60 compared to Geo10 outside of the tropics with similar burden values in this region. The AOD is a measure of turbidity and degradation of sunlight and depends, for sulfate, on the particle size. Small particles scatter more efficiently than larger particles. The effective radius increases from 0.4 μm in Geo10 to 0.45 μm in 10Tg60 (Fig. 9) and, consequently, the AOD decreases. The better representation of the stratospheric dynamics in the higher resolution simulations and the resulting stronger confinement of the particles in the tropics cause them to grow larger.*

Secondly, if the westerly QBO is inhibiting transport out of the tropics, why doesn't this apply to 10Tg30? There are plenty of aerosols in the accumulation mode in the extratropics and the QBO is in an even stronger westerly phase.

In simulation 10Tg30 SO₂ is injected at a level between the lower and the upper branch of the BDC. The plot of particle number of the accumulation mode indicates a transport path in the upper BDC branch. However, concentration of these particles is small and plays only a minor role in the overall picture as shown in the plot of sulfate concentration in the supplementary material.

AOD and burden of 10Tg30 show a strong maximum in the tropics and also the ratio of tropical to extratropical burden values show less meridional transport than Geo10 and 10Tg60.

Besides, if QBOW intensified vertical transport as mentioned earlier in the manuscript, why is the distribution of coarse mode particles in Figure 8 for 10Tg60 (bottom center panel) confined to a smaller vertical region as compared to that in Geo10 (left bottom panel). This plot alone implies higher vertical velocity at the equator in Geo10 as compared to 10Tg60. Again, this difference could be the consequence of different model dynamics, wave breaking and BD circulation due to differences in vertical resolution, and not to the QBO.

It has not been our intention to say that differences between Geo10 and 10Tg60 are only related to the QBO, especially as there is no QBO in 10Tg60. We said in the text: *... to estimate an error in the sulfate forcing made by using a dynamically too simple model....* However, we try to describe this better in the text. We add in the model description and in Section 6 some sentences on the impact of numerical diffusion. Stronger numerical diffusion in vertical direction is the main reason for the broader vertical sulfate layer in Geo10. This is caused by the lower vertical resolution in Geo10.

We add in the model description: *Numerical vertical diffusion is reduced when increasing the vertical resolution, resulting in a lower vertical extend of the sulfate layer.*

We add in the text in Sect 6: *We performed simulations with an injection rate of 10 Tg(S) yr^{-1} in order to enable a direct comparison to the results of (Niemeier and Timmreck, 2015). They*

used ECHAM5-HAM in a 39-layer version that would not simulate an internally generated QBO, but instead constant equatorial easterly winds. This allows us to estimate an error in the sulfate forcing made by using a dynamically too simple model with, additionally, stronger numerical diffusion in vertical direction which present itself as an additional artificial up- and downdraft. However, through the distinction of QBO phases in Section 5 we can clearly attribute effects simulated, in this paper, to changes in the tropical circulation.

Authors state in line 360 that the number density in the coarse mode is lower in Geo10 than in 10Tg60. I don't see that from Figure 8 — that maybe true right at the equator in a very small region, but overall the number density in the coarse mode if anything is bigger in Geo10.

We now show the effective radius in the paper. This indicates better the shift to larger particles. The number density is showing this shift in the changed ratio between accumulation to coarse mode particles.

4. Interpretation of differences in the text between 10Tg60 and 10Tg30 are also not consistent with the figures. For example: Lines 350-351: The difference between 10Tg60 and 10Tg30 is primarily the injection altitude and not the tropical wind system From Figure 10Tg60 shows that likely the aerosol transport out of the tropics occurs via the lower branch of the BDC, where in 10Tg30, the aerosols are transported via mixing and the upper branch of the BDC (see also minor comment 1).

Line 350 to 351 were a very general statement on the differences of burden and AOD in the extratropics. The first paragraph of Section 6.1 changed to: ~~The sulfate burden of the three simulations, given in Figure ??, is similar in the extratropics, while the results of the vertically-integrated aerosol optical depth (AOD) differ between the simulations. So we may assume that the burden is less sensitive to changes in the tropical wind systems than the AOD, which depends not only on the sulfate mass but also on the particle number. The comparison of simulation Geo10, the low resolution model version without internally generated QBO, and simulation 10Tg60 shows in tropics and sub-tropics the impact of the different vertical resolution. The imperfect representation of stratospheric dynamics with constant easterly winds in Geo10 cause a more permeable barrier (Punge et al., 2009) resulting in a stronger meridional transport. The ratio of tropical to extratropical mean burden is lower in Geo10 than in 10Tg60 (Tab. 1), indicating the stronger transport. The model version with lower vertical resolution tends to overestimate meridional transport, a tendency also seen in earlier volcano studies (Niemeier et al. (2009); Timmreck et al. (1999).~~

We also discuss now the transport of accumulation mode particles in 10Tg30 in the upper branch of the BDC in Section 6.1 (see next paragraph) and, additionally, we add some lines to Section 2.1 (see minor comment 1).

Lines 366- 367: 'Figure 8 indicates low meridional transport resulting in low particle number densities in extratropics in 10Tg30.' This is not at all consistent with top right — most panel in Figure 8: there are plenty of particles in the extratropics.

It's true that the text was not very clear. We change the description of the transport at several places in the text. The mentioned paragraph changes to: *Increasing the injection height to 30 hPa, simulation 10Tg30, further intensifies the equatorial confinement, which is stronger at a height of 30 hPa than at a height of 60 hPa. Thus, injection in 10Tg30 results in a strong tropical maxima of burden and AOD, as discussed in Section 5.2. Small particles with little sedimentation are transported vertically in the tropical pipe and meridionally in the upper branch of the BDC while coarse mode particles are transported in the lower branch (Fig 2 supplementary material). In the extratropics the AOD is 30% to 50% lower compared to simulation 10Tg60. Figure ?? indicates low meridional transport, resulting in low particle number densities in the extratropics in 10Tg30. Due to the stronger tropical confinement the particles grow to radii up to 0.75 μm , an increase of about 0.25 μm compared to*

10Tg60. The reduction of AOD in the extratropics is strong enough to reduce the global mean AOD of 10Tg30 compared to 10Tg60 (Tab. 1).

5. In order to explain the differences between AOD in the 3 simulations shown in Figure 7, it would be helpful to plot effective radius of particles and surface area density, instead of what is currently in Figure 8.

We add a figure of effective particle radius to the text and move the figure of particle number density to the supplementary materials.

Minor Comments:

1. Line: 77-78: 'This quasi-horizontal mixing is the main transport branch for sulfate aerosols'. This statement highly depends on the location of injection of the aerosols. There are three main ways the aerosols can be transported out of the tropics: a) The deep branch of the BDC, the shallow branch, and horizontal mixing. Aerosols injected right above the tropical tropopause are mostly going to be transported with the shallow branch of the BDC, those injected several kilometers above the tropopause will likely be primarily transported with the upper branch of the BDC. Some will be transported horizontally by mixing. I suggest that a discussion of the different branches of the BDC is added and how the location of injection (30 hPa and 60 hPa discussed here) affect which branch of the BDC is the primary transport mechanism. Figure 1 of Bonisch et al. 2011 has an excellent graphic (Atmos. Chem. Phys., 11, 3937-3948, 2011 www.atmos-chem-phys.net/11/3937/2011/ doi:10.5194/acp-11-3937-2011) We change the text in Section 2.1: *Sharp gradients of potential vorticity at the edges of the surf zone act as a transport barrier: the polar vortex at high latitudes inhibits transport to the poles in winter months and the equatorial jets of the QBO form, at this edges, the subtropical transport barrier. This subtropical barrier results in the formation of a reservoir for chemical species in the lower tropical stratosphere (Trepte and Hitchman, 1992). The barrier is strongest in heights between 21 km to 28 km (50 to 15 hPa). The strength of the transport barrier depends on the phase of the QBO. These barriers can be seen as 'eddy-transport-barriers' (Mcintyre, 1995). They do not act as barriers to the zonally averaged BDC (Haynes and shuckburgh, 2000). As a consequence, the BDC has two horizontal transport branches, one below and one above the transport barrier. Transport of sulfate out of the tropics occurs mainly in the lower branch of the BDC but for small particles in high level injection scenarios also in the upper branch.*

2. Line 92: An average period of the QBO is 28 (not 29) months.

Done

3. Figure 1: Why isn't the QBO included here for the 4 Tg 30 hPa injection? Please include it.

Done

4. Temperature anomalies in rightmost panel of Figure 2 clearly exceed the colorbar. Please change the colorbar so the maximum and minimum temperature anomalies are clear in all the panels. Please in the text also include the amplitude of maximum temperature anomalies for the simulations in Figure 2.

Done

5. Line 203-204: 'Positive anomaly does not extend to the pole ... because polar vortex blocks the transport' - it would be helpful to overplot the aerosol concentrations here to demonstrate this point clearly.

Done.

6. Why is there such a strong negative temperature anomaly near 5 hPa at the equator in the 8Tg30 hPa simulation? (Rightmost panel of Figure 2)

In the upper stratosphere/lower mesosphere, a significant negative temperature anomaly in the equatorial region and a significant positive temperature anomaly over the extratropics. This has been observed after strong volcanic eruptions.

The diabatic heating of aerosols in the lower stratosphere causes an increase in the residual vertical wind velocity in the tropical pipe and consequently an adiabatic cooling, which dominated the temperature anomaly in the region above the aerosol heating. The residual vertical wind in 10Tg60 and 10Tg30 (Figure 1, supplementary material) is strongest in the region of the strongest vertical wind shear (15 hPa (10Tg60) and 5 hPa (10Tg30), respectively). In this region the adiabatic cooling is strongest (1 K/d and 2 K/d), which corresponds to the height of the temperature minima of both results.

We add in the text: *The temperature anomalies in the upper stratosphere, including the cooling above the heated aerosol layer in the tropics, are caused by the increase of the residual vertical wind and the related adiabatic heating anomalies (Toohey et al., 2014).*

7. Figure 3: The color-scale is inappropriate. It is impossible to see what are the zonal wind velocities in the top panels as well as anomalies in the bottom panels. Both clearly exceed the color scale. Please correct.

Done

8. Figure 8: Again here, the colorbar needs to be adjusted that it is clear what the maximum contour is in the top leftmost panel.

We change the figure to effective radius.

9. Line 445-447: That is too strong of a conclusion! Injecting at 30 hPa and other location could be viable at other latitudes – only equatorial injections have been shown here, hence authors should not make such a sweeping conclusion.

We change the text to: A conclusion from this results can be that injecting at high levels at the equator might be unfavorable for CE, not only because it is technically more demanding.

10. Lines 457- 469: I'm not sure how this paragraph is relevant to the main point of the study. If the authors chose to keep it, please explain how you arrived at the injection estimates up to 2100 mentioned in line 460 and 462.

We change the paragraph to:

In this study we calculate a smaller efficiency of sulfur injections than (Niemeier and Timmreck, 2015) obtained in model simulations with lower vertical resolution and, hence, less realistic tropical dynamics. Therefore we have to modify some of the conclusions drawn in (Niemeier and Timmreck, 2015). They estimated from their simulations an injection of 45 Tg(S) yr^{-1} would counteract global greenhouse gas forcing of 6 W/m^{-2} . This amount would be necessary to keep the global mean temperature at 2020 level in 2100 while maintaining business as usual emissions. The decreased forcing efficiency simulated in this study would increase the injected amount to 70 Tg(S) yr^{-1} , injected at the equator, as lowest estimate to keep 2020 temperature level in 2100. Adapting a strategy of Laakso et al (2017), with injections following the zenith of the sun or injecting at 15 N and 15 S, may slightly reduce the injection rate. However, the spread in the forcing simulated by different model is large (Niemeier and Tilmes, 2017), as is the amount of injected sulfur necessary to generate a certain forcing. Estimates of lifting costs of sulfur into the stratosphere (e.g. Moriyama et al. (2016)) depend strongly on the efficiency of the injection.

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