Reviewer #1

August 21, 2015

We would like to thank reviewer #1 for his/her review. Our answers and the corresponding changes to the manuscript are given below in a blue text colour.

Using an atmosphere-ocean-chemistry climate model, the authors assess the effects of a generic tropical volcanic eruption on stratospheric ozone and the northern hemispheric polar vortex. With a suite of simulation experiments, they separate the ozone effects of the eruption via eruption-related changes in heterogeneous chemistry and stratospheric dynamics, and the feedback of the induced ozone changes on the temperatures and northern hemispheric polar vortex.

General comments:

- This manuscript presents some new interesting results and confirm the findings of previous studies. In this current version the manuscript largely focuses on results that have already been published, such as the comparison between the effects of a Pinatubo-like eruption on the heterogeneous chemistry and dynamics (PD15_HET Vs PD15_RAD). While a reanalyses of previous studies is always useful, I think that this manuscript would gain novelty by focusing on the comparison between present-day and preindustrial, or among the different magnitude of SO2 injections. To my knowledge such a systematic assessment of tropical eruptions in present-day and preindustrial conditions has not been published. While the figures are present (at least for the PD15 and PI15 cases, but not for the 30Tg and 60Tg experiments), they are often rushed in the description. There are nearly no figures with PI30 and PI30.

Thank you for this comment. Unfortunately, it is not possible to show all figures for all the different cases. For Figure 3 the setup of 2 climate states, 3 forcings, and three processes would results in a figure with 18 panels.
In the revised manuscript we therefore added several figures as supplementary material and reference the figures in the manuscript.

- S1: column ozone anomalies for all experiments (present day, similar to Fig. 3)
- S2: column ozone anomalies for all experiments (preindustrial, similar to Fig. 3)
- S3: Zonal mean DJF ozone and residual circulation anomalies in PD15, PD30, PD60, PI15, PI30, and PI60 (similar to Fig 4).

Furthermore, we have added more information on the comparison present day vs. preindustrial to the revised manuscript. Nevertheless, we decided to keep the results on PD15_HET and PD15_RAD since these experiments allow comparing our results to previous studies, which is important for the interpretations of the other experiments, inparticular for the RAD effects.

The contribution of the chemistry-climate interaction is also interesting, even though I am not sure that the title of Section 3.3 is appropriate. I suggest 'Effects of the coupling between ozone and stratospheric dynamics on the stratosphere'.

Thank you for this comment. We changes the title of Section 3.3 using your suggestion.

Aquila et al. (2013), which is included among the references, is a very similar study but limited to the PD15 experiments. I suggest to include more quantitative comparisons with their results, and to extend the conclusions not covered in their study. For instance, Fig. 3 is very similar to Fig. 7 of Aquila et al. (2013). I suggest adding the same figure for PI15, PD/PI30 and PI/PD 60.

Thank you. A discussion of the similarities and differences to Aquila et al (2013) has been added to the discussion section. Furthermore, we include a figure for PI15, PD/PI30 and PI/PD 60 (similar to the Fig. 4) as supplementary material (S3).

"A comparable case-study for the 1991 Mt. Pinatubo was performed by Aquila et al (2013), with a similar separation between radiative-dynamical and heterogeneous chemical effects on the aerosols. They identified a combination of HET-AER and RAD-DYN processes to be responsible for the ozone anomalies in the SH. In particular they found similar anomalies in the residual mean circulation being responsible for reduced ozone in the tropics and enhanced ozone concentrations at mid-latitudes (compare Figure 4b and their Figure 7). This response, however, is limited to the early phase of the eruptions and in combination with the phase of the Brewer-Dobson circulation the anomaly-pattern is found only in the SH, while in the NH ozone anomalies are mainly affected by HET-AER effects. Our findings suggest a similar response of the residual mean circulation in the NH during boreal winter, and we conclude that a combination of RAD-DYN and HET-AER effects are needed to understand ozone anomalies on both hemispheres. This difference in the response is not yet understood, but may be related to differences in the aerosol forcings. Understanding the response to the RAD-DYN mechanism is of particular importance for volcanic eruptions under preindustrial conditions with low load of ozone depleting halogens, where chemical effects become weak and the response is dominated by radiative-dynamical effects. "

- The manuscript is very confusing in the description of the figures. I have found very difficult to follow which figure the authors are describing, and if they are referring to PD or PI.

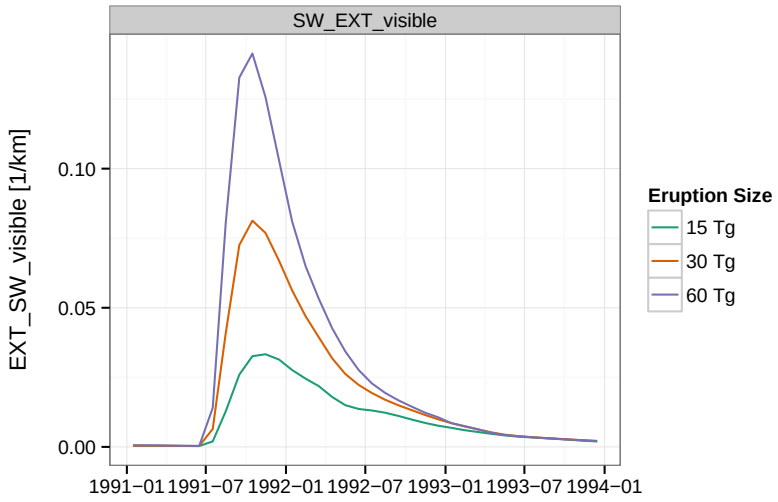

Figure 1: Time series of extintion rates in the visible (440-690nm) at the equator at 54 hPa.

Thank you. In Section 3.1 and 3.2 the describe first changes for the present day climate state and focus on the differences in the preindustrial climate state after that. In the revised manuscript we made the structure clearer by adding subsubsection to these chapters.

- Are aerosol and radiation coupled in AER2D? If not, the dispersal of the aerosol, and therefore the spatial distribution of the forcing, could be totally unrealistic, especially in the case of the 60Tg injections. For instance, a visual comparison of the panels in Fig. 1 suggests that the residence time of the volcanic aerosol is similar for all three injection magnitudes. Is this true, and, if true, is it reasonable? Larger injections should lead to larger particles and faster settling (e.g. English et al., 2012), but also to a larger vertical extent of the volcanic aerosol (Aquila et al., 2014), which would extent the stratospheric lifetime.

Indeed there is no coupling with radiation in AER, which can affect the aerosol distribution. While this is a limitation of our method, the eruptions in this study are idealized, and the radiation-aerosol influence doesn't dominate over the general uncertainties and variations in the transport of the aerosols after an eruption.
The settling effect is present in AER, but is may not be obvious in Fig. 1 of the manuscript. Below (Fig: 1) the extinctions rates in the visible are shown for equatorial latitudes in an altitude of 54 hPa. This figures reveals that about 2 years after the eruption the extinctions are almost independent of the forcing.

Specific comments:

- P14285 L28: PD15 resemble Pinatubo only for the initial conditions (time of the eruption, order of magnitude of the SO2 injected) and GHG scenario, but not in the sense of the initial meteorological conditions and QBO phase (or is the QBO nudged to observations for the period?) nor in the sense of the actual forcing, given that the SAD shown in Fig. 1 does not resemble the

one from SAGE observations, which show that the peak of aerosol was south of the equator. I would rather write that the injection amount and timing of the eruptions are compatible to the eruption of Mt. Pinatubo.

Thank you for this comment. We agree that our present day 15 Tg simulation and the real eruption of Mt. Pinatubo differs in a number of aspects. The QBO, however, which is nugged to observations in the model, does not differ from the 1991 eruption. Also the injection amount may not be high in comparison to Pinatubo. We therefore rewrote the description of the experiment and forcing. In Sect. 2.2 (aerosol forcing), we state now:

"Arfeuille et al (2013) found that an injection of 14 Tg of SO_2 (7 Tg of sulphur) produced mid-visible extinctions much higher than observed in the tropical stratosphere after the Pinatubo eruption. As shown by Dhomse et al. (2014), the peak burden of sulphur in the particle phase was around a factor of two lower than the peak sulphur burden in the gas phase, in the range 3.7 to 6.7 Tg of sulphur. The 15 Tg AER simulation shall therefore be regarded as an upper limit for the perturbation that occurred following Pinatubo. Furthermore, some differences in the shape of the AER aerosol forcing and observations for Pinatubo exists."

And in the experiment section (2.3) we replaced the statement that our PD15 experiment closely resembles the Pinatubo eruption by:

"As explained in section 2.2, the PD15 ensemble simulation represents an upper limit for the effects from the Mt. Pinatubo eruption in 1991."

- P14287 L5: what is the significance level?

Thank you, the significance level is 95%. We have added the necessary information to the end of the Section 2.3:

"Significance estimates are based on a two-tailed Student's t-test using the 5 % significance level."

- P14287 L 21: Cite relevant literature for the chemical mechanism (e.g. Tie and Brasseur (1995) or Granier and Brasseur (1992))

Thank you, the references was added.

- P14287 L 25: Is the reduction of N2O5 by 80% a model result or is it from previous published literature? Adding 'not shown' would help clarify, if it is a model result, otherwise please cite the relative reference.

Thank you. It is a model results and we have added '(not shown)' to the corresponding statement.

- P25399 L10: The authors write that the oscillations in column ozone anomalies are due to polar ozone depletion in the northern and southern hemisphere. However, in Fig. 3b no polar depletion is visible in the southern hemisphere, except for the non-significant depletion in August-September at 60S. Is that negative anomaly what the authors refer to?

Thank you for this comment. The oscillation in column ozone shown in Figure 2 are very small for the 15 Tg eruption but become larger for the 30 and 60 Tg forcing. The spatial pattern of the column ozone anomalies for the 30 and 60 Tg eruption, clearly reveals the amplified polar ozone depletion.
We have included the spatial pattern of column ozone anomalies in the supplementary material (Figures S1 and S2) in the revised manuscript.

- P14288 L13: the polar ozone depletion in RAD is said to increase with forcing strength, but this is not shown in any figure.

The increase in polar ozone depletion from PD15_RAD to PD60_RAD is now shown in Figure S1 and S3 (supplementary material).

- P14289 L11: 'In the following' or 'later' (to indicate the following months)?

Thank you, we changes this to "*In the following months...*".

- P 4289 L16: Do the authors mean Fig. 3h or 3d?

We were referring here to the comparison between PD15 and PD60. In particular Fig 3d is meant here, which we are referring now. Thank you for this question.

- P14289 L25 to L28: This is true for the northern hemisphere, while in the southern hemisphere PD15_RAD and PI15_RAD are not very different from each other. If the reason was the reduced polar ozone depletion, shouldn't the difference between PD and PI be even larger in the southern hemisphere?

Thank you for this question. This is again a difference between present day and preindustrial which is very weak in the 15 Tg experiments, but becomes larger for the larger eruptions. This effect is visible in Figure 2, but only for the global mean column ozone anomalies. In the revised manuscript the column ozone anomalies for all experiments are included as supplementary material Figure S1 and S2. A comparison of S1h (PD60_RAD) to S2h (PI60_RAD) reveals the differences in polar ozone depletion between the two climate states.

- P14290 L11: the warming in PD60 is not shown, correct?

The warming for PD60 is shown in Figure 5d. If you were refering to the PD60_HET simulations: temperature anomalies for this experiment are indeed not shown as figures, but the numbers are mentioned in the manuscript.

Why is there a warming at northern high latitudes in PD15_HET, even though not signifi-

cant?. Is this warming a consistent feature of all ensemble members?

The warming may be related to the NH polar vortex weakening, which is not only present in the ensemble average of PD15_HET but also in the PI15_HET and in the experiments with stronger volcanic forcing. The warming is present in most of the PD15_HET simulations (compare Figure 2, see below). A simple analysis suggest that the relationship between vortex intensity and temperature anomalies in the NH polar stratosphere is robust for the winter season. Figure 3 (see below) displays the relationship between the anomalies in the NH polar vortex intensity (using the u60 index as described in the manuscript) and the temperature anomalies in the polar stratosphere (averaged over the latitudes 75-90N at 40 hPa) for the eight members of the PD15_HET experiment. Both anomalies were calculated relative to the control ensemble average. In the winter after the eruption this relationship is almost linear.
We have added a sentence on the positive temperature anomalies to the revised manuscript:

"The HET-AER effect furthermore causes slight positive temperature anomalies in the NH polar stratosphere (Fig. 5a), which are related to the weakening of the polar vortex. "

- P14290 L16: The black line in Figure 7 is not described anywhere (I suppose it is the average of the reference simulation). Am i suppose to compare the purple line in the upper left panel of Fig. 7 to the black line? If so, u60 is outside of the shaded area only in January and February in the case of PD15.

Thank you. The black line is indeed the control simulations and we add this information to Figure 7. We agree that the u60 index for the HET experiment under present day conditions is significant below the Control in January and February and we will correct this statement in the manuscript. We furthermore, added a monthly mean comparison for January and March to Figure 7 and discuss the differences between the experiments for mid- (January) and late-winter (March) in the results section.

"The weakening of this $\bar{u}60$ index due to the HET-AER effect is mainly a phenomena of the mid to late winter (January, February). In January the vortex intensity reduces to $35\pm15\,m/s$ in the PD15_HET-AER experiment in comparison to $48\pm11\,m/s$ in the CTRL experiment (32 ± 18 and $36\pm11\,m/s$ for the 30 and 60 Tg experiment,respectively. Compare Fig. 7). During spring a slight, but not significant vortex intensification is found for the stronger forced ensemble simulations. In March mean value of the vortex intensity in CTRL is $9\pm18\,m/s$, while the vortex in PD15_HET-AER reaches an average of $14\pm15\,m/s$ (21 ± 19 and $19\pm13\,m/s$ for the 30 and 60 Tg experiment,respectively). "

- P14290 L25: Is the temperature anomaly in PD30 shown anywhere? If not, how do we know that it is linear? What do the author mean with 'the temperature response seems to saturate' in the PD60 case? Is it because it is not equal to three times the PD15 temperature response? The upper limit of the color scale is not indicated, so it is difficult to understand if the temperature response really saturates.

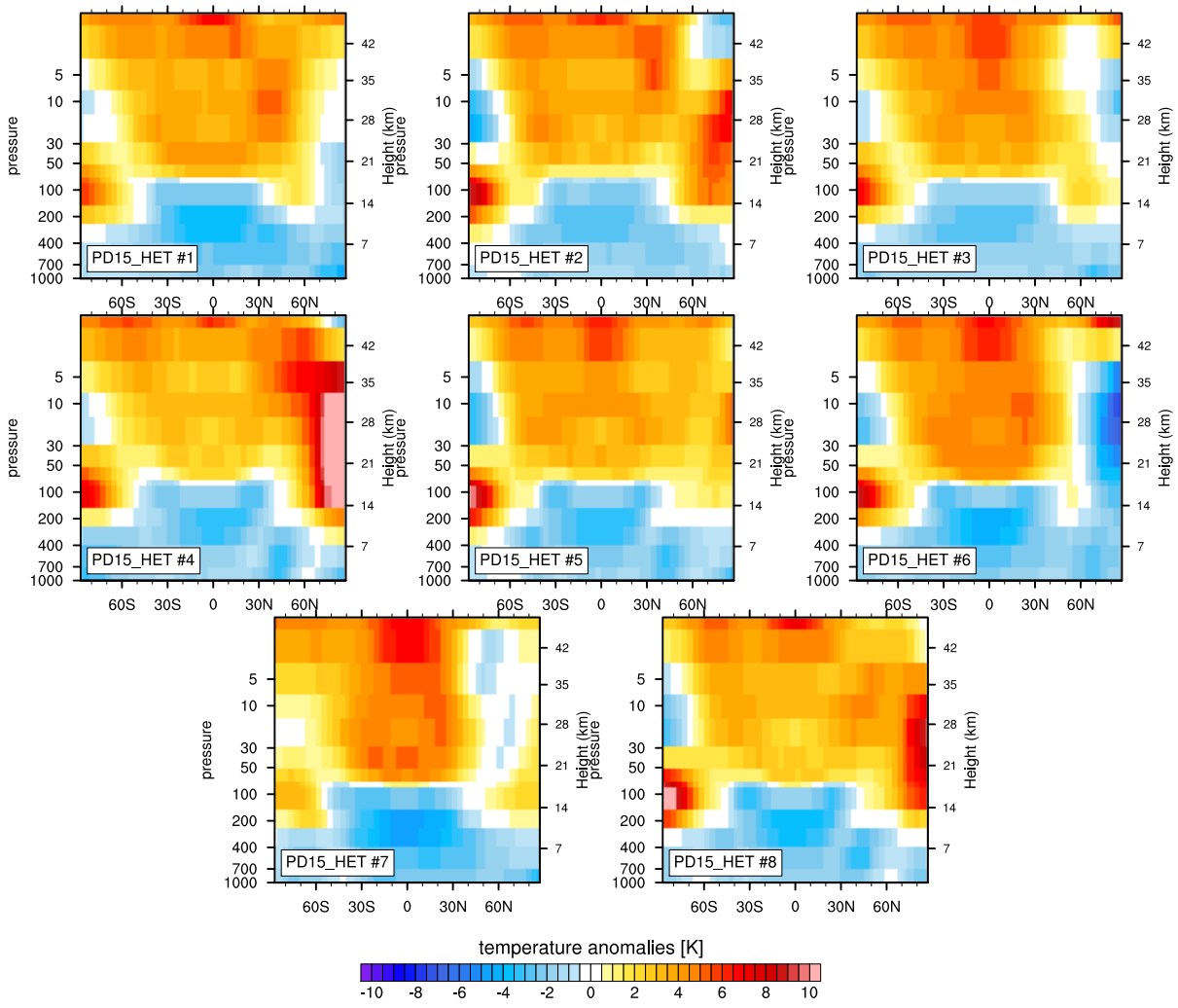

Figure 2: DJF temperature anomaly in the 8 ensemble members from the PD15_HET experiment (similar to Fig.5 of the ACPD manuscript). Anomalies were calculated relative to the ensemble average of the PD control simulations.

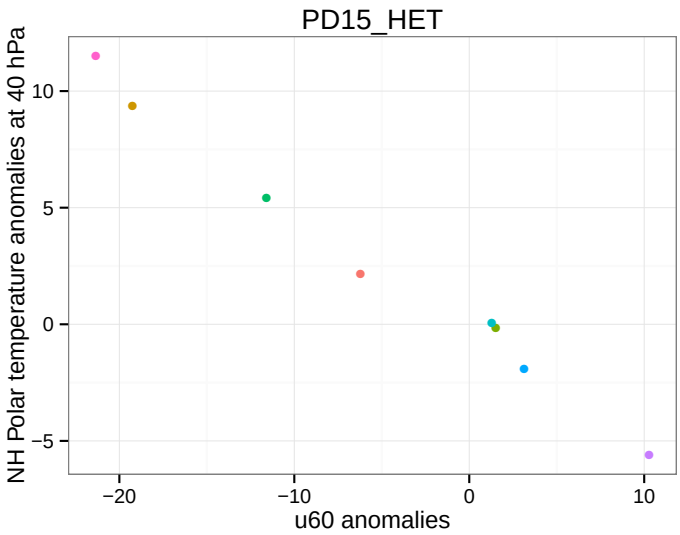

Figure 3: Scatterplot of (x-axis) the NH polar vortex anomalies and (y-axis) temperature anomalies in the polar stratosphere for the first post-eruption DJF season in the individual members of PD15_HET.

The statement of an almost linear temperature increase from PD15 to PD30 is based in the analysis of the model results, but figures for the PD30 temperature anomaly are not shown. In the revised manuscript we give numbers for the maximum temperature anomalies for all forcings:

"As expected, the temperature anomalies increase with rising aerosol mass. At 50 hPa the maximum temperature anomaly, which occurs around December, is 9.5 K for the 15 Tg and increases to 18.2 and 21.7 K for the 30 and 60 Tg eruptions, respectively."

- P14291 L7: To which figure do these lines refer to?

Thank we, we have added references to the Figures to the revised manuscript.

"The amplitude of the temperature change through the HET-AER mechanism is much weaker than the changes caused by the RAD-DYN effect (Fig. 5a). Nevertheless, the temperature reduction causes a significant weakening of the NH polar vortex, but only a slight increase in the vortex intensity is found for the RAD-DYN experiment for the 15 Tg aerosol forcing (Fig. 7)."

- P14291 L111: how does the different patterns of the temperature anomaly exactly translates into a different dynamical response?

Our hypothesis is that the difference between a cooling which is limited to tropical latitudes (HET) and a warming which is present at almost all latitudes, is responsible for the different dynamic response. However, with the current setup, we have no possibility to test this hypothesis. We rewrote this paragraph to make this more clear:

"The difference in the response of the NH polar vortex is not yet fully understood. It may be related to the different patterns of the temperature anomaly. The aerosol induced warming covers all latitudes up to 60 N in the lower and middle stratosphere and reaches even polar latitudes in the upper stratosphere. By contrast, the cooling associated with the HET-AER effect is limited to the SH and up to 30° N due to the seasonal cycle of the Brewer-Dobson circulation."

- P14291 L23: The comparison between Fig. 5c and Fig. 5g is difficult, I would add a third line with difference plots.

Thank you. We have added the temperature differences between present day and preindustrial for the different experiments to Figure 5 and reference them on several occasions in the revised manuscript.

- P14296 L6: a comparison with observations is not very significant, since the forcing itself of Fig. 1 does not look like the observed aerosol distribution. However, I agree that the reason is probably the excessive warming of the lower stratosphere.

Thank you. We are aware of the fact that we can expect some differences between our simulations and the observations caused only the the differences in the forcing. This is why the state at the beginning of this paragraph:

"A direct comparison of our results to observations is difficult given the highly idealised character of our experiments."

Are brominated very-short lived substances included? Oman et al. (2014) shows that it could enhance ozone depletion.

Yes, we have added to the model two brominated very-short lived substances recommended by Liang et al., (2010).

- Liang, Q., R. S. Stolarski, S. R. Kawa, J. E. Nielsen, J. M. Rodriguez, D. R. Blake, E. L. Atlas, and L. E. Ott (2010), Finding the missing stratospheric Bry: A global modeling study of CHBr3 and CH2Br2, Atmos. Chem. Phys., 10, 2269-2286.

Refenreces

- English, J. M., Toon, O. B., and Mills, M. J. (2012). Microphysical simulations of sulfur burdens from stratospheric sulfur geoengineering. Atmospheric Chemistry and Physics, 12(10), 4775–4793. http://doi.org/10.5194/acp-12-4775-2012

- Aquila, V., Garfinkel, C. I., Newman, P. A., Oman, L. D., and Waugh, D. W. (2014). Modifications of the quasi-biennial oscillation by a geoengineering perturbation of the stratospheric aerosol layer. Geophysical Research Letters, 41. http://doi.org/10.1002/(ISSN)1944-8007

- Aquila, V., Oman, L. D., Stolarski, R., Douglass, A. R., and Newman, P. A. (2013). The Response of Ozone and Nitrogen Dioxide to the Eruption of Mt. Pinatubo at Southern and Northern Midlatitudes. Journal of Atmospheric Science, 70(3), 894–900. http://doi.org/10.1175/JAS-D-12-0143.1

- Tie, X., and Brasseur, G. (1995). The response of stratospheric ozone to volcanic eruptions: Sensitivity to atmospheric chlorine loading. Geophysical Research Letters, 22(22), 3035–3038. http://doi.org/10.1029/95GL03057

- Granier, C., and Brasseur, G. (1992). Impact of heterogeneous chemistry on model prediction of ozone changes. Journal of Geophysical Research, 97(D16), 18015– 18033.

- Oman, L. D., and Douglass, A. R. (2014). Improvements in Total Column Ozone in GEOSCCM and Comparisons with a New Ozone Depleting Substances Scenario. Journal of Geophysical Research-Atmospheres, 119, 5613–5624. http://doi.org/10.1002/2014JD021590

Reviewer #2

30. August 2015

This paper presents an interesting analysis of the changes in stratospheric ozone and polar vortex characteristics which would occur for idealised major volcanic eruptions in present-day and pre-industrial conditions, separately quantifying the two major pathways that the enhanced stratospheric aerosol perturbs stratospheric ozone, via heterogeneous chemical reactions on the aerosol surfaces, and by dynamically-induced changes associated with the radiative heating of the volcanic aerosol layer. The paper will be of great interest as this is, to my knowledge, the first time the two path ways have been so comprehensively assessed in the present-day and pre-industrial setting. Furthermore the sensitivity experiments injecting larger amounts of sulphur (approximately twice and four times larger injection Pinatubo) and assessing the ozone and polar vortex responses are particularly interesting.

Thank you for your this very helpful and exceptional detailed review! Our answers and the resulting changes to the manuscript, are given below.

Summary and General Comments:

The paper is generally well written with the results section and Figures presenting the findings in a logical and thoughtful way, with the discussion and conclusions then summarizing the main findings with appropriate explanations and references. However, the Introduction and Abstract are a little clumsily worded in places and require some improvement. I have therefore made rather a large number of suggested minor changes which should be made before publication.

Thank you for the very detailed comments and suggestions. We applied almost all of them and made substantial changes to the abstract.

My first general comment here concerns the way the different effects from the eruption are being categorized. The Introduction (beginning pg 14278, line 18) explains each of the different ways that the injected sulphur from a tropical eruption can perturb stratospheric ozone chemistry. Then later in the Introduction these effects are classified or grouped into two different types: radiative effects and chemical effects. The latter is explained to only refer to heterogeneous chemical effects from the aerosol itself, whereas chemical effects due to modified PSC occurrence are included in the radiative effect classification, since the PSC changes result from the radiative-dynamical effects of the volcanically enhanced aerosol.

My general comment here is that I would strongly recommend that the authors clarify the way the 'chemical effect' is presented in the Title, Abstract and Conclusions.

The title of the article refers to separating radiative from chemical effects but it needs to be made clear that when you say chemical you just mean the 'direct chemical effects' of the enhanced aerosol. I consider that it is actually rather a nice approach being taken in the article to quantify the indirect chemical effects of the enhanced stratospheric sulphur and consider relative magnitude compared to the direct chemical effects.

As such I suggest that the authors consider slightly changing the wording of the title to ensure the nature of the separation is immediately clear.

I suggest to change:

'The impact of volcanic aerosols on stratospheric ozone and the Northern Hemisphere polar vortex: separating radiative from chemical effects under different climate conditions'.

to

'The impacts of volcanic aerosol on stratospheric ozone and the Northern Hemisphere polar vortex: separating radiative-dynamical changes from direct effects due to enhanced aerosol heterogeneous chemistry.'

Thank you for this comment. The suggested title is indeed more precice and we adopted you version for the revised version of the manuscript. For changes to the abstract and conclusions see below.

This leads to my second major comment which is that throughout the text the indirect impacts are referred to as 'radiative effects'. That is confusing because there are of course radiative effects from the direct chemical effects of the enhanced aerosol as well as those from the dynamical/photolysis changes. To clarify I suggest the authors replace all sinstances of 'radiative effects' with 'radiative-dynamical effects' and replace the three-letter-acronym 'RAD' with the more descriptive 'RAD-DYN' Similarly the heterogeneous chemical effects isolated via the HET experiment is just the effect of greater heterogeneous chemistry occurring on the volcanically-enhanced aerosol surfacea (not accounting for any changes due to modified PSC occurrence). So I recommend also to change 'HET' to 'HET-AER'. That way it is clear that you're not including in that the effects from the enhanced or modified PSCs.

Thank you for this comment. We replaced the acronym RAD by the suggested RAD-DYN and HET by HET-AER throughout the manuscript. Furthermore, we clearified the definition of the two groups by the following statement in the introduction sections:

"Please note, that the RAD-DYN effect includes also chemical effects, for instance through changes in the reactions rates or PSC formation due to the temperature changes. Moreover, HET-AER processes affect also the radiation transfer through the atmosphere by changes in the chemical composition."

Another general comment is that at several points in the text the wording 'climate states' is used. The reader may expect the 'climate states' to refer to a large ensemble of experiments e.g. carried out over a range of El Nino and/or North Atlantic Oscillation conditions at the time of the eruption, whereas in fact the authors are only referring here to the differences in greenhouse gas and ozone depleting substances in pre-industrial and present-day simulations.

The setup of the ensemble simulations was performed in a way that a large range of different ENSO and AMOC states was considered. Eight simulations are probably not enough to talk about a 'large ensemble of experiments', but at least we can reduce some imprint of internal variability on the results.

The word 'states' should not be used in conjunction with 'climate' as that implies some additional analysis wherby simulations have been stratified and grouped to try understand the effect of different ENSO or NAO state on the response. That is not what is done here. The word 'setting' is more appropriate and I have pointed to instances where this should be changed in the Specific Comments below. Similarly the word 'climate' is used but in fact much of the effects focus on different halogen loading, so the effects are really composition-climate rather than just climate. I therefore recommend to replace 'climate conditions' with 'composition-climate setting'. Note this is also just singular for setting because there is only one different setting considered (pre-industrial vs present-day).

Thank you for this comment. We revised the manuscript accordingly, removed the term 'climate state' from the manuscript and replaced it either by 'climate setting'. However, we decided to keep the term 'preindustrial/present day conditions'. Although we focus mainly on the influence of different ODS concentrations, this can not be separated from the influence of the different GHG concentrations and their effect in the mean climate. We think the term 'preindustrial/present day conditions' is therefore more appropriate, since it covers all differences between these two periods.
We were not completely confident with the term 'composition-climate setting' and therefore decided to clarify the role of the composition and their effect on the climate in the Introduction:

"Moreover, we assess the influence of the eruption strength on these changes and the role of different climate setting in moderating the dynamical responses, where the term climate setting describes a specific atmospheric composition of greenhouse gases and their effects on the climate system."

... and in the 'Experiment' Section:

"To assess the role of the climate setting on the response, the eruptions either take place under present day (early 1990s with high loads of ODS and GHG in the atmosphere) or preindustrial conditions (early 19th century, low concentrations of ozone depleting halogens and GHG). The simulations therefore differ in their atmospheric composition, but also the climate state is different due to the effect of the GHG on temperature and dynamics."

My final general comment is that the Abstract, particularly the 2nd half, is of poor quality, whereas the results and discussion are clearly of high quality. I think that the 1st author may have mistakenly worded the last few sentences there (see Specific Comments 7 and 8). I have made suggestions (comments 1 to 6) to improve the Abstract but the last part requires a re-write for the authors to express what they intended to say here.

See below for the changes to abstract.

The end of the Conclusions section was also suprisingly weak and the statements need to be made more quantitative. The size of the effects are mostly quantified in the results section but need to be cited much more clearly for the author to get an overall idea of how significant or otherwise are the changes being discussed. This requires changes to the results, conclusions and Abstract.

See below for the corresponding changes to the abstract and the conclusions.

Overall the manuscript represents a valuable scientific contribution and I recommend publication once the revisions and comments have been addressed. However, the shortcomings in the 2nd half of the Abstract and the conclusions are sufficient that the revisions required are such that I consider them major and would like to see the revised version again before the article be allowed to proceed to publication.

Specific Comments:

1) Title – As above suggest to change
'The impact of volcanic aerosols on stratospheric ozone and the Northern Hemisphere polar vortex: separating radiative from chemical effects under different climate conditions'.
to
'The impacts of volcanic aerosol on stratospheric ozone and the Northern Hemisphere polar vortex: separating radiative-dynamical changes from direct effects due to enhanced aerosol heterogeneous chemistry under different composition-climate setting.'

Thanks again, the title was changed

2) Abstract – pg 14277, lines 2-4: The first sentence seemed strangely worded with the 'are modulated by' not really appropriate in this context. Suggest to simplify the 'After strong volcanic eruptions stratospheric ozone changes are modulated by heterogeneous chemical reactions (HET) and dynamical perturbations related to radiative heating in the lower stratosphere (RAD)' with 'After major volcanic eruptions the enhanced aerosol causes ozone changes due to greater heterogeneous chemistry on the particle surfaces (HET-AER) and from dynamical effects related to the radiative heating of the lower stratosphere (RAD-DYN).'

Thank you. we changed the first sentence as suggested

3) Abstract – pg 14277, lines 4-7: Again the wording seemed a little clumsy here with too much specifics mentioned in that one sentence. There is no need to mention the specific model used in the abstract. I suggest to reword the current text from: 'Here, we assess the relative importance of both processes as well as the effect of the resulting ozone changes on the dynamics using ensemble simulations with the atmosphere-ocean-chemistry-climate model (AOCCM) SOCOL-MPIOM forced by eruptions with different strength' to 'We carry out a series of experiments with an atmosphere-ocean-chemistry-climate model to assess how these two processes change stratospheric ozone and polar vortex dynamics.'

Thanks. We modified the second sentence based on your suggestion.

"We carry out a series of experiments with an atmosphere-ocean-chemistry-climate model to assess how these two processes change stratospheric ozone and NH polar vortex dynamics."

4) Abstract – pg 14277, lines 7-9: Following on from above, suggest to include some of the specifics from that above sentence into this follow-on sentence. Suggest to replace
'The simulations are performed under present day and preindustrial conditions to investigate changes in the response behaviour'
with
'Ensemble simulations are performed under present day and preindustrial conditions, and with aerosol forcings representative of different eruption strength, to investigate changes in the response behaviour.'

Again, thank you. We applied the suggested changes.

5) Abstract – pg 14277, lines 9-10: You say here that the HET effect is only relevant under present day conditions. That is not quite true because although the halogen induced ozone loss is near zero in the pre-industrial setting, the N2O5 hydrolysis component of HET is still occurring and influences Arctic stratospheric ozone (as you show in Fig 3e). As well as changing the wording to clarify this, the sentence could also be re-worded slightly to improve the Abstract readability. I'd suggest therefore to replace:
'The results show that the HET effect is only relevant under present day conditions and causes a global reduction of column ozone'
with something like this:
'We show that whereas the halogen component of the HET effect dominates under present day conditions, globally reducing column ozone particularly at high latitudes, in a preindustrial atmosphere the HET effect increases stratospheric ozone due to N2O5 hydrolysis.'

Thank you. We changed the sentence following suggestion:

" We show that the halogen component of the HET-AER effect dominates under present day conditions with a global reduction of ozone (peak reduction -21 DU for the strongest eruptions) particularly at high latitudes, whereas the HET-AER effect increases stratospheric ozone due to N2O5 hydrolysis in a preindustrial atmosphere (maximum anomalies +4 DU). "

6) Abstract – pg 14277, lines 11-12: You say 'These ozone changes' but you need to be specific that you're referring to the ozone changes due to the aerosol heterogeneous chemistry and also better to put in context of the overall strengthening of the polar vortex caused by the radiativedynamical changes. Suggest to replace:
'These ozone changes further lead to a slight weakening of the Northern Hemisphere (NH) polar vortex during mid-winter.'
with
'The halogen-induced ozone changes in the present-day atmosphere offset part of the strengt-

hening of the Northern Hemisphere (NH) polar vortex during the first posteruption winter.'

Thanks, the suggested change was applied and the sentence reads now:

" The halogen-induced ozone changes in the present-day at- mosphere offset part of the strengthening of the NH polar vortex during mid-winter (reduc- tion of up to -16 m/s in January) and slightly amplify the dynamical changes in the polar stratosphere in late winter (+11 m/s in March)."

6) Abstract – pg 14277, lines 12-13 – replace 'climate state' with 'composition-climate setting'.

We rewrote this sentence to:

" The RAD-DYN mechanism leads to positive column ozone anomalies which are reduced in a present day atmosphere by amplified polar ozone depletion (maximum anomalies +12 and +18 DU for present day and preindustrial, respectively). "

7) Abstract – pg 14277, lines 14-18 – I don't think this is correct. I can only assume you are referring here to the HET effect not the RAD effect.

In this case we are indeed refering to the RAD-DYN effect, which lead to a colder polar stratosphere, more PSC and more clorine activation on the PSCs under present day conditions. As visible, for instance, in Fig. 2 the RAD-DYN timeseries differ between preindustrial and present day in the way, that under in a present day atmosphere the positive anomalies weaken at the end of the SH and NH winter season. This is discussed at pg 14288 line 5ff of the submitted manuscript.
In the revised manuscript we shown now column ozone anomalies for all experiments as supplementary material (Fig. S1).

8) Abstract – pg 14277, lines 18-21 – this needs re-writing with much more quantitative statement about the relative magnitude of the effects. The authors have designed experiments to isolate these effects and yet the current summary of the findings is not adequate.

We have added a number of quantitative statement to the abstract (see changes above).
We also rewrote the last sentences of the manuscript:

"For preindustrial conditions, the ozone response is consequently dominated by RAD-DYN processes, while under present day conditions, HET-AER effects dominate. The dynamical response of the stratosphere is dominated by the RAD-DYN mechanism showing an intensification of the NH polar vortex in winter (up to +10 m/s in January). Ozone changes due to the RAD-DYN mechanism slightly reduce the response of the polar vortex after the eruption under present day conditions."

9) Introduction – pg 14277, lines 23-25 – sentence improved by deleting 'volcanic', ', which are', 'gases' and 'can' – more succinct and easier to read.

Thank you. That is indeed a much better sentence:

"Tropical eruptions strong enough to inject into the stratosphere perturb the physical and the chemical states of the climate system for several years and longer."

10) Introduction – pg 14277, line 25-26 – replace 'Among the large number of eruption products...' with 'Although a range of gases are injected (e.g. Textor et al., 2004)....' and insert 'global climate impacts stem from the aerosol produced from the injected' between 'the' and 'sulphur dioxide' then delete 'has probably the strongest climate impact'.

Thank you. The sentence was modified to

"Although a range of gases are injected (e.g. Textor et al., 2004), the global climate impacts stem from the aerosol produced from the injected sulphur dioxide (SO2)."

11) Introduction – pg 14278, line 1 – insert ', volcanically injected' after 'the stratosphere' and before 'SO2....'

done.

12) Introduction – pg 14278, line 5 – replace 'The aerosols increase' with 'the enhanced stratospheric aerosol increases'.

done, thank you.

13) Introduction – pg 14278, lines 7-8 – replace 'The absorption of long wave radiation' with 'Increased absorption of long wave and solar near-infra-red radiation', replace 'aerosol cloud' with 'volcanic plume', replace 'which leads to' with 'causing' and rplace 'in these regions' with 'of the tropical stratosphere'.

The sentence was changed to

"Increased absorption of long wave and solar near-infrared radiation increases heating rates in the volcanic plume causing a pronounced warming of the tropical stratosphere."

14) Introduction – pg 14278, lines 8-10 – replace 'affect' with 'alter' and replace 'by interaction' with 'via interactions' and replace 'even the climate at the surface' with 'affect surface climate'.

done.

"The perturbed vertical and meridional temperature gradients alter the stratospheric circulation and via interaction between the stratosphere and the troposphere affect surface climate."

15) Introduction – pg 14278, lines 10-12 – delete 'that has been'.

ok.

16) Introduction – pg 14278, lines 14-16 – replace 'Anomalous positive surface temperatures' with 'Such surface temperature anomalies', replace 'the coupling of the' with 'interactions between the' and replace the 'the' before 'trospheric circulation' and insert 'patterns' afterwards.

applied, thank you.

17) Introduction – pg 14278, paragraph beginning line 18 needs re-wording to make it easier for the reader to digest. I think the follow changes help...

Line 18 replace 'effect' with 'overall impact', delete 'the' and 'further'. Line 19 insert 'the effects from' before '(i)' then reword that point replacing 'the effect of the changing temperature on the reaction rates' with 'altered reaction rates due to changes in temperature'.

Line 20 for point (ii) replace 'the heterogeneous chemistry on the sulphuric acid aerosols' with 'enhanced heterogeneous chemistry from elevated sulphuric acid aerosol surface area density (SAD)',

Line 21 for point (iii) replace 'the effect of the temperature changes and the aerosols on the polar stratospheric clouds (PSC)' with 'the temperature and aerosol changes in modifying the occurence and types of polar stratospheric clouds (PSCs).'

Line 22 for point iv) insert 'composition' before 'changes induced' to distinguish this from the temperaturechanges mentioned in points i) and iii).

Line 23 for point v) delete 'the' and insert 'from the enhanced aerosol' after 'photolysis rates'.

Thank you. all suggested changes were applied in the revised manuscript.

18) Introduction – pg 14278, lines 24-28 – reduce these 2 sentences merging into one replacing 'In particular, the' with 'The', deleting the text 'is of importance. This reactive effectively', replace 'with the effect of' with ',' and delete 'where the NOx cycle dominates the depletion'.

The suggested changes were applied.

19) Introduction – pg 14278 line 28-29 and pg 14279 lines 1-3. This long sentence repeats partly the sentence before I therefore suggest to remove most of the last 3 lines on that pg 14279 replacing the overall sentence with: 'In the lower stratosphere, the Clx and HOx cycles are more important with the net chemical effect being ozone loss in the present day atmosphere (give a suitable reference here perhaps the recent SPARC ozone assessment?).'

Thank you. The suggested modification was applied.

"In the lower stratosphere, the Clx and HOx cycles are more important with the net chemical effect being ozone loss in the present day atmosphere (Tie and Brasseur, 1995; Solomon et al., 1996; SPARC, 2013). "

20) Introduction – pg 14279 lines 3-5 – merge these 2 sentences and reword as 'The chemical ozone loss from an eruption in the present-day atmosphere is intensified at high latitudes by a

strenghtening of the polar vortex, which reduces temperatures, increasing PSC occurrence.

thank you. We corrected this sentence following your suggestion.

21) Introduction – pg 14279 lines 5 to 7 – replace 'Secondly' with 'Additionally, ' and insert ', in combination with the colder temperatures,' after 'polar stratosphere', replace 'an additional type of PSC' with 'liquid sulphuric acid ternary solution particles' replace 'surfaces for' with 'surface area density and therefore', replace 'reactions' with 'ozone loss' and replace 'on PSCs' with (Carslaw et al., 1994).

thank you. We corrected this sentence following your suggestion.

22) Introduction – pg 14279 line 11 – delete 'are expected to'.

applied.

23) Introduction – pg 14279 lines 12-15 – replace 'With important quantities of additional anthropogenic ozone depleting halogens in the atmosphere the net...' with 'In the present day atmosphere, the elevated halogen loading in the stratosphere means that the net chemical...'.

Thank you, we changed the sentence accordingly.

24) Introduction – pg 14279 line 16 – replace 'reactions are expected to' with 'effect of the eruption is to'.

Thank you, we changed the sentence accordingly.

25) Introduction – pg 14279 lines 21-22 – replace 'The effect' with 'These effects' and delete 'of a tropical eruption' (that's implied) and then replace 'therefore be roughly divided into two processes' with 'broadly be classified into two distinct groups'.

Done.

26) Introduction – pg 14279 lines 22-26 – this seems to long-winded to me. Suggest to reduce this passage to 'The first involves composition-dynamical interactions associated with the radiative absorption of the volcanic aerosol, which we refer to as RAD-DYN.'

Thank you. We applied the suggested changes.

27) Introduction – pg 14279 lines 26-30 – similarly improve wording at the start by replacing 'The second process includes a large...' with 'The second comprises the net change due to a large...'

Thank you. We applied the suggested changes.

28) Introduction – pg 14279 line 30 and pg 14280 lines 1-2 – delete this sentence and simply add at theend of theprevious one ', which we refer to as HET-AER.'

Thank you. We applied the suggested changes.

29) Introduction – pg 14280 line 3 – replace 'observations it is difficult' with 'observations alone it is not possible' and delete the comma after 'understand'.

Thank you.

30) Introduction – pg 14280 line 9 insert 'observed to ' before 'reduce' and give appropriate reference.

We have include Grant et al. (1994) and Randel et al. (1995) as references:

"In the tropics, total column ozone was observed to reduced after the eruption of Mt. Pinatubo (Randel et al. 1995) which was a combined signal of a reduction in the lower stratosphere and an increase of ozone concentrations above (Grant et al. 1994). "

31) Introduction – pg 14280 line 14 replace 'is responsible for an increase in the ozone' with 'via chemical'.

Thanks, suggested change was applied.

32) Introduction – pg 14280 line 15-16 replace 'dominated' with 'dominates', replace 'leading to positive ozone anomalies' with 'increasing ozone' and add at the end of that sentence ', with further enhancement in the tropics due to modified photolysis (Pitari and Rizi, 1993).'

Thank you, we applied the suggested modifications.

33) Introduction – pg 14280, lines 16-17 – with the above change (32) can then delete this sentence beginning 'Furthermore'

Sentence was deleted.

34) Introduction – pg 14280, lines 20-27 – the sentences in this paragraph seem out of order. Start with the current last sentence (describing the NH ozone changes) and replace 'The ozone changes' with 'Ozone changes...' and add 'thought to be' before 'primarily' Then start a 2nd sentence 'By contrast, the increasing ozone observed in the SH has been attributed....' adding also Dhomse et al. (2015) after the Aquila reference.

Thank for this comment. The paragraph was changed to:

"Ozone changes in the NH are thought to be primarily caused by the heterogeneous chemical reaction effects (Pitari and Rizi, 1993; Aquila et al., 2013), in particular at high latitudes (Portmann et al., 1996; Solomon et al., 1996; Rosenfield et al., 1997; Telford et al., 2009; Pitari et al., 2014). By contrast, the increasing ozone observed in the Southern Hemisphere (SH) has been attributed to dynamical processes induced by the aerosol heating in combination with the phase of the Brewer–Dobson circulation (Aquila et al., 2013; Dhomse et al., 2014) or the Quasi-Biennial-Oscillation (QBO) (Telford et al., 2009; Randel and Wu, 1995)."

35) Introduction – pg 14281, line 16 – replace 'different climate states' with 'different composition-climate setting' (singular because you only show one different setting).

We changed this to:

"Moreover, we assess the influence of the eruption strength on these changes and the role of different climate setting in moderating the dynamical responses, where the term climate setting describes a specific atmospheric composition of greenhouse gases and their effects on the climate system."

36) Section 2.2, pg 14283, lines 8-9 – delete these as you have already said this in the Introduction.

Sentence was deleted

37) Section 2.2, pg 14283, lines 23-24 – You say the stratospheric warming various more or less linearly with the SO2 mass injected. I don't think that is the case. Since the warming is asscociated with the near-infra-red and longwave absorption this is mainly driven by the way the coarse particles evolve. I would expect the increase in the coarser particle abundance with great SO2 injection to be even more non-linear than the accumulation mode sizes. So why do you expect the strat-warming to scale linearly? Please give a reference or replace with a setence that states that you expect the warming to be even more non-linear than the SAD changes.

The absorption efficiency does not change largely in the range of sizes where most of the aerosol mass is present after an eruption (Grainger et al., 1995). The stratospheric warming varies thus mostly linearly with the aerosol volume, but some deviations can occur, also due to the part of the absorption in the near-infrared and because of the decreased residence time of large aerosols.

- Grainger, R.G., Lambert, A., Rodgers, C.D., Taylor, F.W.: Stratospheric aerosol effective radius, surface area and volume estimated from infrared measurements, Journal of Geophysical Research, 1995.

38) Section 2.2 pg 14284 line 15 – please join up this para with the previous 1st sentence – it

shouldn't start a new paragraph here.

Done.

39) Section 2.2 pg 14284 line 18 – replace 'was much lower' with 'was around a factor of two lower' and insert 'the peak sulphur burden' before 'in the gas phase'.

Thank you, changes were applied .

40) Section 2.2 pg 14284 line 20 – replace 'a stronger' with 'an upper limit for the'.

Thank you, changes were applied .

41) Section 2.3 pg 14285 lines 3, 5, 17, 21 and 23 – replace 'climate state' with 'composition-climate setting'.

We rewrote this paragraph using either 'climate setting' or 'preindustrial/present day conditions.

42) Section 2.3 pg 14285 line 14 – you have referred to the 'Atlantic Meridional Overturning Circulation'. Is that what you mean here? Or do you rather mean the North Atlantic Oscillation (NAO)?

We are referring to the AMOC. All simulations were initialized at 1. of January. The eruption take place in the mid of June. The state of the NAO about half a year prior to the eruption is probably not an important factor and controlling the state of the NAO at the beginning of the eruption is not possible with this setup. However, given the slow adjustment time of the ocean, the state of the AMOC or ENSO may be important and therefore the ensemble was initialized in a way that a large number of different AMOC/ENSO states are covered.

43) Section 2.3 pg 14285 line 26 – replace '. Therefore the model was forced with' with ', based on'.

Thank you, the sentence was modified accordingly

44) Section 2.3 pg 14285 line 27 – insert (after AER model.) 'Note that in this study we do not include the effects of the enhanced aerosol in reducing photolysis, and related composition changes'.

Thank you, the sentence was modified accordingly

45) section 2.3 pg 14285 line 27 – replace 'Note that the' with 'As explained in section 2.2., the....' and replace 'closely resembles the conditions of' with 'represents an upper limit for the effects from'

Thank you, the sentence was modified accordingly

46) section 2.3 pg 14285 line 28 – replace 'Moreover, the PI60' with 'The PI60'

done.

47) section 2.3 pg 14286 line 6 – replace 'radiative perturbations (RAD)' with 'radiativedynamical perturbations (RAD-DYN)' and replace 'reactions (HET)' with 'reactions on aerosol surfaces (HET-AER)'.

done.

48) section 2.3 pg 14286 line 7 replace '(RAD)' with '(RAD-DYN)' and in all other parts of the text and Figures.

We replaced RAD by RAD-DYN throughout the manuscript.

49) section 2.3 pg 14286 line 8 replace '(HET)' with '(HET-AER)' and in all other parts of the text and Figures.

We replaced HET by HET-AER throughout the manuscript.

50) section 2.3 pg 14286 line 10 – add 'with 8 members (Table 1)' after 'Ensemble experiments...'

Thank you, the suggested modification was applied.

51) section 2.3 pg 14286 line 18 replace 'show the pure effect' with 'isolate only the'

Thank you. Sentence was changed.

52) section 2.3 pg 14286 line 26 add 'based on 95% confidence interval'.

Thank you. We changed this sentence to:

" Significance estimates are based on a two-tailed Student's t-test using the 5 % significance level."

53) section 3.1 pg 14287 lines 10-12. Insert 'As expected, ' at the start of this sentence then replace 'anomalies reveals amplified ozone depletion' with 'anomalies (Fig 3a) shows largest ozone depletion' and replace 'during the winter months (Fig 3a)' with 'during spring'.

Thank you. The sentence was changed as suggested.

54) section 3.1 pg 14287 line 13. Move '(Fig 4a)' from the end of the sentence

Done.

55) section 3.1 pg 14287 line 14. Insert 'aerosol heterogeneous' before 'chemical effect'.

Thank you

56) section 3.1 pg 14287 line 24. Insert '(not shown)' after '30 hPa' and replace '<' with 'around'.

Thank you

57) section 3.1 pg 14287 line 27. Replace 'reductions of ozone' with 'reductions in ozone (Fig 3a)'.

Done.

58) section 3.1 pg 14288 line 11 Suggest to insert 'due to additional PSC occurrence in the stronger colder polar vortex' after 'experiment'.

In HET-AER the vortex is not stronger/colder in comparison to RAD-DYN, therefore this explanation for the stronger polar ozone anomalies in HET-AER may be missleading. We decided to keep this sentence unchanged.

59) section 3.1 pg 14288 line 13 Insert '(not shown)' at the end of the sentence.

Done.

60) section 3.1 pg 14288 line 20 Insert '(Fig 3b)' at the end of the sentence.

Done.

61) section 3.1 pg 14289 line 1 Insert '(Fig 3b)' at the end of the sentence.

Done.

62) section 3.1 pg 14289 line 8 Replace 'radiative effects' with 'radiative-dynamical effects'

Done.

63) section 3.1 pg 14289 lines 15-16 replace '(Fig. 3e and d)' with '(Fig 3d)'.

Done.

64) section 3.1 pg 14289 line 29 Replace 'radiative effect' with 'radiative-dynamical effects'

Done.

65) section 3.2 pg 14290 line 5 Replace 'the question how ozone changes' with 'to ask how the ozone changes'

Done.

66) section 3.2 pg 14289 line 8 insert 'aerosol' before 'direct radiative effect'.

Done.

Just to clarify – does the size of the warming effect you're citing for the RAD-DYN include also the offset from any dynamical changes in ozone and water vapour? It would seem that this is included or are you just isolating the aerosol radiative heating here? Can you quantify each individual contribution from the simulations you have done?

The size of the RAD-DYN warming includes the aerosol radiative heating and dynamical changes in water vapour and ozone. Quantifying the contribution from the dynamical ozone changes on the temperature changes was done on section 3.3, where we force an ensemble of simulations with the ozone changes simulated in RAD-DYN (compare also Figure 9). These results show that ozone changes cause a slight warming and therefore amplify the aerosol radiative heating. Quantifying the warming from dynamical water vapour changes is not possible with the current setup.

67) section 3.2 pg 14289 line 21 replace '(RAD)' with '(RAD-DYN)'.

Done.

68) section 3.2 pg 14290 line 1 insert 'the' before 'case of the'.

OK.

69) section 3.2 pg 14290 line 2 replace 'In contrast' with 'By contrast'.

OK.

70) section 3.2 pg 14290 line 2 replace 'Contrary' with 'By contrast'.

OK.

71) section 3.2 pg 14290 line 11 replace '<' with '>'. Also consider adding 'due to the seasonal cycle of the Brewer Dobson circulation'.

The cooling is limited to the SH and tropical latitudes, therefore '<' seems to be correct. However, we have modified this sentence and added the suggest explanation using the BDC.

"By contrast, the cooling associated with the HET-AER effect is limited to the SH and up to 30° N due to the seasonal cycle of the Brewer-Dobson circulation."

72) section 3.2 pg 14290 lines 15-16 I don't see this effect. The red and green lines are suprisingly similar – please look again and consider changing the 'is clearly visible' statement....

With the sentence "In particular the NH polar vortex weakening in mid-winter due to the HET-AER effect is clearly visible in the u60 index" we are refering to the blue line in Fig 7 (HET). This index shows a significant weakening in mid winter (dots at the bottom), which we consider as 'clearly visible'.
In the revised manuscript we furthermore added subfigures to Fig 7 showing the monthly mean statistics for January and March for each experiment. In these figures, the vortex weakening in the HET-AER experiment is also very clear.

73) section 3.2 pg 14290 lines 22-24 You say the tropical stratospheric warming is weaker in the preindustrial atmosphere. But I don't see this from Figures 5c and 5g. Perhaps it is the colour scale that doesn't show contrast between 6 and 8K warming (for example). Please can you explicitly state the maximum warming values in each case here.

The differences is small for the 15 Tg eruption and therefore not well visible with the selected colour scale in Fig 5c and 5g. For larger eruptions the temperature difference between preindustrial and present day increases as can be seen in Fig 5d and 5h (in this case for the full forcing experiments). In the revised manuscript we quantify the differences between preindustrial and present day for each eruption size:

"RAD-DYN effects in a present day atmosphere slightly differ from the response under present day conditions. At 50 hPa the maximum tropical stratospheric warming is 1.2 K larger in the present day atmosphere and this differences increases to 2.3 and 1.7 K for the 30 and 60 Tg eruption. The stronger warming under present day conditions is not ..."

74) section 3.2 pg 14290 lines 23-30 In this sentence you say the warming is stronger under pre-industrial conditions. This seems to clash with the previous statement (see

Thank you, this was wrong. present day conditions were meant here. We corrected this in the revised manuscript.

73)? Please clarify what you mean here. Also I wonder about the explanation given below. Can't this just be explained by the weaker aerosol heterogeneous chemical ozone loss?

We found significant differences in the stratospheric warming between the present day and preindustrial RAD-DYN experiments. The heterogeneous chemial effect is not considered in theses experiments. Consequently, the differences in the tropical stratospheric heating can only be related to (a) direct radiative effects, (b) dynamical changes in ozone or water vapour. Dynamical ozone change were found not to responsible, as shown in section 3.3 and Figure 9b and 9f. Furthermore, there is no indication that water vapour may explain the differences (however, we con not directly estimate the effect of dynamical water vapour changes in the current setup). Therefore, we argue that the radiative effects are responsible to the temperature differences.

75) section 4 pg 14294 line 14 insert 'aerosol heterogeneous' before 'chemical effect'.

Applied.

76) section 4 pg 14294 line 27 replace 'Contrary' with 'By contrast'

Applied.

77) section 4 pg 14295 line 1 insert 'sub-tropical and mid-latitude' before 'lower stratosphere'.

Applied.

78) section 4 pg 14295 line 4 replace 'climate state' with 'composition-climate setting'.

We replaced 'climate state' by 'atmospheric composition'.

79) section 4 pg 14295 lines 19-20 move comma from after 'larger' to instead be after 'present day'.

Thank you.

80) section 4 pg 14295 lines 24-25 replace 'is characterised by very similar boundary conditions as' with 'can be considered similar to the perturbation from'

Thank you, we applied the suggest modification.

81) section 4 pg 14296 line 3 insert 'sub-tropics and ' before 'mid-latitudes'.

Done.

82) section 4 pg 14296 line 18 replace 'leads' with 'lead'.

Done.

83) section 4 pg 14296 line 19 delete comma after 'considered'.

Done.

84) section 4 pg 14297 line 2 replace 'Furthermore, the' with 'The', insert 'also' after 'study'.

Done.

85) section 4 pg 14297 line 3 replace 'an aerosol' with 'a 2D global aerosol'

Done.

86) section 4 pg 14297 line 13 delete 'for'.

Done.

87) section 4 pg 14297 lines 12-15 – I thought this was a weak end to a good article. Please can you look again and see whether you have the numbers here to quantify the proportion of each effect in terms of the peak ozone changes with some numbers e.g. for the global changes shown in Figure 2. Also for the peak temperature changes in the tropics (or other latitude bands) can you quote what fraction comes from HET-AER and what fraction from RAD-DYN?

Thank you for this comment. We rewrote the last paragraph of the conclusion section:

"In summary, we show that ozone is affected globally by a volcanic eruption for several years. Both effects, the radiative dynamical perturbation by the volcanic aerosols as well as heterogeneous chemical reaction on the aerosols are important for the response of the ozone chemistry. The climate setting, in particular the atmospheric concentrations of ODS, has the strongest effects on the heterogeneous chemical effect on aerosol surfaces with pronounced global ozone depletion for present day ODS concentrations (peak reductions of -13, -18, and -21 DU for the 15, 30, and 60 Tg eruptions, respectively) and slight ozone increase for preindustrial ODS concentrations (between 4–5 DU for all eruptions). Radiative dynamical ozone changes are positive for preindustrial and present day conditions, but for present day the response is weakened by amplified polar ozone depletion (+5,+8,+12 DU peak column ozone anomalies for present day and +6, +14, +18 DU for preindustrial). The full effect of the volcanic aerosol, therefore, clearly differs between preindustrial and present day, with long lasting ozone depletion in a present day atmosphere and positive ozone anomalies for preindustrial conditions. The response of stratospheric temperature and dynamics is dominated by the radiative heating effect of the aerosols. A small influence of the climate setting on the heating of the lower tropical stratosphere was found, with larger temperature anomalies for the present day experiments. Dynamical radiative ozone changes further amplify the stratospheric temperature anomalies in the lower tropical stratosphere (and cause a cooling in higher levels). Ozone changes due to heterogeneous chemical reactions on the aerosols are responsible for a slight cooling of the tropical stratosphere. In winter and early spring after the eruption, the NH polar vortex is intensified, due to the radiative warming in the tropical stratosphere. Ozone changes, either due to radiative-dynamical effects or heterogeneous reaction on the aerosol surface, induce a slight weakening of the vortex in mid-winter. In late winter they cause a slight strengthening of the westerly circulation in the NH polar stratosphere."

88) Figure 2 caption pg 14307 insert 'heterogeneous aerosol' before 'chemical effect' and change 'HET' to 'HET-AERO' and also in Figure. Similarly please change 'radiative aerosol effects' to 'radiative-dynamical aerosol effects' and replace 'RAD'with 'RAD-DYN' in caption and in Figure.

Thank you. The figure caption was modified accordingly.

89) Figure 3 caption pg 14308 replace 'columns' with 'column' and delete redundant text 'between January of the eruption year (year 0) and 40 months after the eruption'. Also replace 'Anomalies are calculated relatively to the corresponding control ensemble mean and the stippling in the simulation panels...' with 'Stippling...'.

Thank you. The figure caption was modified accordingly.

90) Figure 4 caption pg 14309 insert 'aerosol' before 'heterogeneous chemical'.

Thank you. The figure caption was modified accordingly.

91) Figure 5 caption pg 14310 insert 'aerosol' before 'heterogeneous chemical' and replace 'HET' with 'HET-AER'.

Thank you. The figure caption was modified accordingly.

References:

- Carslaw, K. S., B. P. Luo, S. L. Clegg, Th. Peter, P. Brimblecombe and P. J. Crutzen (1994). Stratospheric aerosol growth and HNO3 gas phase depletion from coupled HNO3 and water uptake by liquid particles, Geophys. Res. Lett., vol. 21, no. 23, pp. 2479–2482.
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- Textor, C., H.-F. Graf, C. Timmreck and A. Robock (2004). Emissions from volcanoes, chapter 7 in 'Emissions of Atmospheric Trace Compounds' Ed: Granier, C., Artaxo, P., Reeves, C. E.

Thank you, the references are included in the revised manuscript.

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The impacts of volcanic aerosol on stratospheric ozone and the Northern Hemisphere polar vortex: separating radiative-dynamical changes from direct effects due to enhanced aerosol heterogeneous chemistry

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Abstract

After ~~strong volcanic eruptions stratospheric ozone changes are modulated by heterogeneous chemical reactions (HET) and dynamical perturbations~~ major volcanic eruptions the enhanced aerosol causes ozone changes due to greater heterogeneous chemistry on the particle surfaces (HET-AER) and from dynamical effects related to the radiative heating ~~in~~ of the lower stratosphere (~~RAD). Here, we assess the relative importance of both processes as well as the effect of the resulting ozone changes on the dynamics using ensemble simulations with the atmosphere–ocean–chemistry–climate model (AOCCM) SOCOL-MPIOM forced by eruptions with different strength. The~~ RAD-DYN). We carry out a series of experiments with an atmosphere-ocean-chemistry-climate model to assess how these two processes change stratospheric ozone and Northern Hemispheric (NH) polar vortex dynamics. Ensemble simulations are performed under present day and preindustrial conditions, and with aerosol forcings representative of different eruption strength, to investigate changes in the response behaviour. ~~The results~~ We show that the ~~HET effect is only relevant~~ halogen component of the HET-AER effect dominates under present day conditions ~~and causes apronounced~~ with a global reduction of ~~column ozone . These ozone changes further lead to a slight weakening of the Northern Hemisphere (NH)~~ ozone (-21 DU for the strongest eruption) particularly at high latitudes, whereas the HET-AER effect increases stratospheric ozone due to N_2O_5 hydrolysis in a preindustrial atmosphere (maximum anomalies +4 DU). The halogen-induced ozone changes in the present-day atmosphere offset part of the strengthening of the NH polar vortex during mid-winter ~~. Independent from the climate state the RAD mechanism changes the column ozone pattern with negative anomalies in the tropics and positive anomalies in~~ (reduction of up to -16 m/s in January) and slightly amplify the dynamical changes in the ~~mid-latitudes. The influence of the climate state on the RAD mechanism significantly differs in the polar latitudes, where an amplified ozone depletion during the winter months is simulated~~ polar stratosphere in late winter (+11 m/s in March). The RAD-DYN mechanism leads to positive column ozone anomalies which are reduced in a present day atmosphere by amplified polar

ozone depletion (maximum anomalies +12 and +18 DU for present day and preindustrial, respectively). For preindustrial conditions, the ozone response is consequently dominated by RAD-DYN processes, while under present day conditions~~. This is in contrast to the preindustrial state showing a positive column ozone response also in the polar area.~~ , HET-AER effects dominate. The dynamical response of the stratosphere is ~~clearly~~ dominated by the ~~RAD~~ RAD-DYN mechanism showing an intensification of the NH polar vortex in winter ~~. Still under present day conditions ozone~~ (up to +10 m/s in January). Ozone changes due to the ~~RAD~~ RAD-DYN mechanism slightly reduce the response of the polar vortex after the eruption under present day conditions.

1 Introduction

Tropical ~~volcanic eruptions , which are~~ eruptions strong enough to inject ~~gases~~ into the stratosphere ~~, can~~ perturb the physical and the chemical states of the climate system for several years and longer (Robock, 2000; Cole-Dai, 2010; Timmreck, 2012). ~~Among the large number of eruption products, the~~ Although a range of gases are injected (e.g. Textor et al., 2004) , the global climate impacts stem from the aerosol produced from the injected sulphur dioxide (SO_2)~~has probably the strongest climate impact~~. In the stratosphere volcanically injected SO_2 is converted into sulphuric acid ($H_2SO_4 + H_2O$) aerosols that (i) reflect in the visible part of the solar spectrum, (ii) absorb terrestrial and solar infrared radiation, and (iii) provide surface for a large number of chemical reactions that alter the chemical composition of the stratosphere (Forster et al., 2007). The ~~aerosols increase~~ enhanced stratospheric aerosol increases the optical depth of the atmosphere, leading to a decrease in SW radiation in the troposphere and at the surface. ~~The~~ Increased absorption of long wave ~~radiation by the aerosols~~ and solar near-infrared radiation increases heating rates in the ~~aerosol cloud, which leads to pronounced warming in these regions~~volcanic plume causing a pronounced warming of the tropical stratosphere. The perturbed vertical and meridional temperature gradients ~~affect~~ alter the stratospheric circulation and ~~by~~ via interaction between the stratosphere and the troposphere ~~even the climate at the surface~~

affect surface climate. A prominent example for this mechanism is the winter warming pattern in the NH ~~that has been~~ observed after several large tropical volcanic eruptions (Robock and Mao, 1992; Stenchikov et al., 2002; Shindell et al., 2004; Fischer et al., 2007; Christiansen, 2008; Zanchettin et al., 2012). ~~Anomalous positive surface temperatures~~ Such surface temperature anomalies over Eurasia are related to a positive phase of the Arctic Oscillation, which is forced by ~~the coupling of the~~ interactions between the stratospheric polar vortex and ~~the tropospheric circulation~~ tropospheric circulation patterns (Graf et al., 1993; Kodera, 1994).

The ~~effect~~ overall impact of a tropical eruption on ~~the~~ stratospheric ozone chemistry can be ~~further separated into~~ separated into the effect from (i) ~~the effect of the changing temperature on the reaction rates~~ altered reaction rates due to change in temperature, (ii) ~~the heterogeneous chemistry on the sulphuric acid aerosols~~enhanced heterogeneous chemistry from elevated sulphuric acid aerosol surface area density (SAD), (iii) the ~~effect of the temperature changes and the aerosols on the~~ temperature and aerosol changes in modifying the occurence and types of polar stratospheric clouds (~~PSC~~PSCs), (iv) the composition changes induced by the dynamical perturbations in the stratosphere, and (v) changes in ~~the photolysis rates~~ photolysis rates from the enhanced aerosol. The temperature change and the reactions on the heterogeneous aerosol surfaces mainly take place in the aerosol cloud. ~~In particular, the~~ The heterogeneous conversion of nitrogen oxides (N_2O_5) into nitric acid (HNO_3) ~~is of importance. This reaction effectively~~ slows down the ~~NO_x~~ NO_x cycle of catalytic ozone destruction, with the effect of increasing ozone concentrations in the middle stratosphere ~~, where the NO_x cycle dominates the depletion~~ (Tie and Brasseur, 1995; Solomon et al., 1996). In the lower stratosphere, ~~where~~ the Cl_x and HO_x cycles are more important ~~, the net-effect is a reduction of the ozone abundance, since the slow-down of the NO_x cycle and heterogeneous reactions intensify the and cycles (Tie and Brasseur, 1995; Solomon et al., 1996) . In polar areas, the eruption is expected to intensify the ozone depletion. Firstly, with a stronger polar vortexthe air temperature inside the vortex is reduced, better isolated from the mid latitudes, and more PSCs can be formed. Secondly~~with the net chemical effect being ozone loss in the present day atmosphere

(Tie and Brasseur, 1995; Solomon et al., 1996; SPARC, 2013) . The chemical ozone loss from an eruption in the present-day atmosphere is intensified at high latitudes by a strengthening of the polar vortex, which reduces temperatures, increasing PSC occurrence. Additionally, the presence of H_2SO_4 in the polar stratosphere in combination with colder temperatures facilitates the formation of ~~an additional type of PSC~~liquid sulphuric acid ternary solution particles, which further ~~increases the surfaces for heterogeneous reactions on PSCs.~~ increase SAD and therefore ozone loss (Carslaw et al., 1994) .

The net effect of the chemical response further depends on the background composition of the atmosphere. The slow-down of the NO_x cycle and heterogeneous reactions ~~are expected to~~ intensify the chlorine cycle of ozone destruction, but chlorine levels have undergone serious changes in the last decades (Solomon, 1999). ~~With important quantities of additional anthropogenic ozone depleting halogens in the atmosphere the net~~ In the present day atmosphere, the elevated halogen loading in the stratosphere means that the net chemical effect of the eruption on the global ozone abundance is a reduction (Tie and Brasseur, 1995; Rozanov et al., 2002; Austin et al., 2013). For low halogen loadings, however, the chemical ~~reactions are expected~~ effect of the eruption is to increase ozone globally (Tie and Brasseur, 1995; Anet et al., 2013; Austin et al., 2013). Furthermore, increase in the stratospheric water vapour concentrations associated with the warming of the tropical tropopause can accelerate the HO_x cycle and reduce ozone even in the case of low halogen concentrations (Heckendorn et al., 2009).

~~The effect of a tropical eruption~~ These effects on stratospheric ozone can ~~therefore be roughly divided into two processes~~broadly be classified into two distinct groups. The first ~~process involves changes in the radiation transfer through the atmosphere with the main effect of increased heating rates in the tropical lower stratosphere. This leads to pronounced changes in the dynamics, which change the meridional and vertical distribution of ozone. Furthermore, in regions with high optical depth the reduced incoming UV radiation can induce ozone depletion. The second process includes~~ involves composition-dynamical interactions associated with the radiative absorption of the volcanic aerosol, which we refer to as RAD-DYN. The second comprises the net change due to a large number of

heterogeneous chemical reactions on the aerosol surface, which ~~change the chemical composition of the stratosphere and strengthen or weaken different catalytic cycles of ozone destruction. In the following the first process refers to the radiative effect of aerosols on the ozone chemistry (RAD), while the second process is named the chemical effect of aerosols (HET)~~we refer to as HET-AER. Please note, that the RAD-DYN effect includes also chemical effects, for instance though changes in the reactions rates or PSC formation due to the temperature changes. Moreover, HET-AER processes affect also the radiation transfer through the atmosphere by changes in the chemical composition.

From the observations it is ~~difficult to understand ,~~ not possible to understand which processes are responsible for the ozone changes and how these changes affect the dynamics. Using model simulations, the attribution of the ozone changes after the Mt. Pinatubo eruption has been assessed in several studies (Brasseur and Granier, 1992; Pitari and Rizi, 1993; Al-Saadi et al., 2001; Telford et al., 2009; Aquila et al., 2013). In general, ~~they~~ these studies conclude that both mechanisms are important, with regional and seasonal differences.

In the tropics, total column ozone was observed to reduced after the eruption of Mt. Pinatubo (Randel and Wu, 1995) which was a combined signal of a reduction in the lower stratosphere and an increase of ozone concentrations above (Grant et al., 1994) . The reduction in the lower stratosphere has been attributed to a mix of the dynamic effect and heterogeneous chemical reactions (Rosenfield et al., 1997; Rozanov et al., 2002). The former reduces ozone by enhanced up-welling of ozone poor air and the latter ~~is responsible for an increase in the ozone~~ via chemical depletion by active chlorine. Above 18 hPa the NO_x deactivation ~~dominated, leading to positive ozone anomalies. Furthermore, Pitari and Rizi (1993) identified similar anomalies~~ dominates, increasing ozone with further enhancement in the tropics ~~related to changes in the UV radiation.~~ due to modified photolysis (Pitari and Rizi, 1993) . A negative feedback between the tropical ozone changes and aerosol heating has been suggested by Al-Saadi et al. (2001), with an reduced heating of up to 25 % in the tropical middle stratosphere.

~~The differences in the ozone response between the NH and Southern Hemisphere (SH), i.e. a reduction in the NH but increasing ozone concentrations in the SH,~~ Ozone changes in the NH are thought to be primarily caused by the heterogeneous chemical reaction effects (Pitari and Rizi, 1993; Aquila et al., 2013) , in particular at high latitudes (Portmann et al., 1996; Solomon et al., 1996; Rosenfield et al., 1997; Telford et al., 2009; Pitari et
By contrast, the increasing ozone observed in the Southern Hemisphere (SH) has been attributed to dynamical processes induced by the aerosol heating in combination with the phase of the Brewer–Dobson circulation ~~(Aquila et al., 2013)~~ (Aquila et al., 2013; Dhomse et al., 2014) or the Quasi-Biennial-Oscillation (QBO) (Telford et al., 2009; Randel and Wu, 1995). ~~The ozone changes in the NH are primarily caused by the heterogeneous chemical reaction effects (Pitari and Rizi, 1993; Aquila et al., 2013) , in particular at high latitudes (Portmann et al., 1996; Solomon et al., 1996; Rosenfield et al., 1997; Telford et al., 2009; Pitari et~~

The stratospheric dynamic perturbation after volcanic eruptions originates mainly from the aerosol heating in the tropical lower stratosphere. Nevertheless, changes in the ozone concentrations also affect heating rates and therefore modulate the dynamic response to the eruption (Muthers et al., 2014a). Using observed ozone anomalies for the Mt. Pinatubo eruption, Stenchikov et al. (2002) found a strengthening of the Arctic Oscillation (AO) in late winter and early spring after the eruption, which is explained by the cooling effect of the polar stratospheric ozone depletion. However, by forcing the model with observed ozone anomalies, a separation of the dynamical and chemical causes of the ozone changes is not possible. Similar results were found by Shindell et al. (2003) who compared the Mt. Pinatubo eruption in simulations with and without ozone changes. For a different climate state (Mt. Tambora, 1815) without anthropogenic chlorine in the stratosphere, they found a very small influence of the ozone changes on the dynamical perturbation.

The purpose of this study is to deepen our understanding of the processes which drive ozone changes after a strong tropical volcanic eruption and how these changes modulate atmospheric dynamics in the stratosphere~~and troposphere~~. Moreover, we assess the in-

fluence of the eruption strength on these changes and the role of different climate ~~states~~ setting in moderating the dynamical responses, where the term climate setting describes a specific atmospheric composition of greenhouse gases and their effects on the climate system. To the best of our knowledge these questions have not been addressed before. Here, we use a set of ensemble sensitivity simulations performed by the ~~AOCCM~~ atmosphere-ocean-chemistry-circulation model (AOCCM) SOCOL-MPIOM. To evaluate the dynamic response this study focuses mainly on the NH and on the winter season.

The paper is structured as follows: Sect. 2 introduces the coupled ~~atmosphere–chemistry–ocean circulation model~~AOCCM, the forcing data, and the setup of the experiments. The results are presented in Sect. 3, first for the simulated ozone changes associated with the different processes (Sect. 3.1), followed by the dynamical changes (Sect. 3.2). Section 3.3 focuses on interactions between the ozone chemistry and the dynamic response associated with the ~~RAD~~ RAD-DYN effects. Finally, we discuss the results and present conclusive remarks in Sect. 4.

2 Model and experiments

2.1 SOCOL–MPIOM

SOCOL-MPIOM is a coupled atmosphere–ocean–chemistry–climate model (Muthers et al., 2014b). The atmospheric-chemistry component SOCOL version 3 (Schraner et al., 2008; Stenke et al., 2013) is based on the physical component MA-ECHAM5 (Roeckner et al., 2003; Manzini et al., 2006), which is coupled to the chemistry module MEZON (Rozanov et al., 1999; Egorova et al., 2003). The chemistry module uses temperature fields from ECHAM5 and calculates the tendency of 41 gas species, taking into account 200 gas-phase, 16 heterogeneous, and 35 photolytical reactions. Heterogeneous reactions are parametrised following Carslaw et al. (1995) and can take place in/on aqueous sulphuric acid aerosols and on three types of polar stratospheric clouds.

In the short-wave scheme of SOCOL the solar spectrum is divided into six spectral intervals. The scheme considers Rayleigh scattering, scattering on aerosols and clouds, and the absorption of solar irradiance in UV-induced photolysis reactions, by O_3, O_2, and 44 other species. In the near-infrared intervals absorption by water vapour, CO_2, N_2O, CH_4, and O_3 is implemented. Furthermore, a parametrisation for the absorption of radiation by O_2 and O_3 in the Lyman-alpha, Schumann-Runge, Hartley and Higgins bands is implemented following an approach similar to Egorova et al. (2004). The long-wave scheme considers frequencies from 10–3000 cm^{-1} for the absorption by water vapour, CO_2, O_3, N_2O, CH_4, CFC-11, CFC-12, CFC-22, aerosols, and clouds. Chemistry–climate interactions can optionally be disabled in the model, by deactivating the interactive chemistry module. In this case three dimensional time dependent ozone data needs to be applied as forcing.

The spectral truncation used in this study is T31, which corresponds to a horizontal resolution of approximately $3.75^\circ \times 3.75^\circ$. In the vertical, the atmosphere is divided into 39 levels with the highest level at 0.01 hPa (80 km). With this vertical resolution the model is not able to produce a ~~Quasi-Biennial Oscillation (QBO)~~ QBO by itself, therefore a QBO nudging is applied (Giorgetta et al., 1999).

The atmosphere-chemistry model SOCOL is coupled to the ocean model MPIOM which includes a sea ice module (Marsland, 2003; Jungclaus et al., 2006). MPIOM is used in a nominal resolution of 3°, with the poles shifted to Greenland and Antarctica to avoid numerical singularities at the poles. This setup allows for a high resolution in the deep water formation region of the North Atlantic. Both models, MPIOM and SOCOL are coupled by the OASIS3 coupler (Budich et al., 2010; Valcke, 2013).

2.2 Aerosol forcing data sets

~~In this study we assess the influence of radiative and chemical effects of stratospheric aerosols on stratospheric ozone chemistry and dynamics for different eruption sizes.~~ To simulate the climatic effect of a volcanic eruption in SOCOL-MPIOM, the model needs information about the optical properties of the aerosols, including extinction coefficients, single scattering albedo, and the asymmetry factor for each spectral interval, latitude, vertical level,

and time step. Furthermore, the chemistry module needs the surface area density (SAD) of the aerosols. In a sensitivity study for different eruption sizes, these forcings needs to be generated in a consistent manner, to allow for a fair comparison between the eruptions.

Since nucleation, condensation, coagulation, and sedimentation of the aerosols change in different ways with the SO_2 concentration (Timmreck et al., 2010), a simple linear scaling of ~~observation based~~ observation-based aerosol data sets by the sulphur mass may lead to an unrealistic forcing data set. The aerosol coagulation process, for instance, depends on the SO_2 concentration and hence particles tend to be larger as we inject more SO_2. The increase in total SAD is hence not proportional to the increase in SO_2 mass. Conversely, the total stratospheric warming depends on the aerosol absorption in the infrared and varies more or less linearly with the SO_2 mass injected (Grainger et al., 1995) . However, increase in the sedimentation rates with larger aerosols further modifies the relationship between the stratospheric warming and the initial sulphur mass released.

The aerosol data set used here, was therefore calculated offline using the micro-physical aerosol model AER (Weisenstein et al., 1997, 2007). AER is a 2-D model with global domain, resolving the atmosphere from the surface to approximately 60 km altitude. The vertical resolution is about 1.2 km with a horizontal grid spacing of 9.5° of latitude. To simulate the formation of the aerosols the SO_2 injection mass, as well as the timing and latitude/altitude of the injection are used as inputs for the AER model. Three AER simulations were carried out with 15, 30, and 60 Tg of SO_2 injected, the former corresponding approximately to the SO_2 detected in the stratosphere shortly after the Pinatubo eruption (Guo et al., 2004). Furthermore, the timing (mid of June) and the location (5° S–14° N between 23–25 km) are chosen to fit the Pinatubo 1991 eruption. Besides the sulphur mass, the same set of initial and boundary conditions were applied in all three simulations, corresponding to the atmospheric state at the time of the eruption of Mt. Pinatubo in 1991 (Fleming et al., 1999).

SAD values and extinction coefficients in the visible are shown in Fig. 1 for the DJF season in the first winter after the eruption on the left and as Hovmoeller diagram for SAD and extinctions at 50 hPa on the right.

~~Note that~~ Arfeuille et al. (2013) found that an injection of 14 Tg of SO_2 (7 Tg of sulphur) produced mid-visible extinctions much higher than observed in the tropical stratosphere after the Pinatubo eruption. As shown by Dhomse et al. (2014), the peak burden of sulphur in the particle phase was ~~much lower than~~ around a factor of two lower than the peak sulphur burden in the gas phase, in the range 3.7 to 6.7 Tg of sulphur. The 15 Tg AER simulation ~~should~~ shall therefore be regarded as ~~giving a stronger perturbation than~~ an upper limit for the perturbation that occurred following Pinatubo. Furthermore, some differences in the shape of the AER aerosol forcing and observations for Pinatubo exists. Using satellite based aerosol forcings, the agreement with observations ~~could~~ can be improved for Pinatubo (Arfeuille et al., 2013). However, due to the non-linear relationship between sulphur mass and the aerosol properties ~~the latter could not be used~~ satellite based aerosol records can not be applied in this sensitivity study. A detailed comparison of satellite based aerosol data sets and the AER method is given in Arfeuille et al. (2013).

2.3 Experiments

A number of ensemble sensitivity experiments were performed. The experiments differ in the aerosol forcing, the ~~climate state, and~~ atmospheric composition of different ozone depleting substances (ODS) and other greenhouse gases (GHG), as well as the physical and chemical processes considered.

To assess the role of the climate ~~state~~ setting on the response, the eruptions either take place under present day ~~conditions~~ (early 1990s with high loads of ~~ozone depleting substances (ODS) and greenhouse gases (GHG)~~ ODS and GHG in the atmosphere) or preindustrial conditions (early 19th century, low concentrations of ozone depleting halogens and GHG). ~~Initial conditions for the two climate states~~ The simulations therefore differ in their atmospheric composition, but also the climate state is different due to the effect of the GHG on temperature and dynamics. The initial states for the two periods are based on two transient simulations described in Muthers et al. (2014b). For the preindustrial climate ~~state~~ simulations the restart files were selected between the years 1812 to 1814. Present day initial conditions were extracted between 1988 to 1990. Each ensemble consists of

8 simulations and the restart files were carefully selected to cover a wide range of different ~~states~~ phases of the Atlantic Meridional Overturning Circulation and the tropical Pacific (ENSO). The timing of the eruption (mid of June) is identical for both climate ~~states~~settings.

For comparison, the simulated response of the climate system after the volcanic eruption is evaluated against an ensemble of control simulations for each climate ~~state~~setting. These simulations were initialised using the same set of initial conditions, but were forced by an aerosol data set representing the unperturbed background state of the atmosphere.

To distinguish between the aerosol forcings and the climate ~~states~~ setting the simulations are named PI15, PI30, and PI60 for the eruption sizes of 15, 30, and 60 Tg of SO_2 in the preindustrial ~~climate state~~conditions, respectively. The present day simulations are named PD15, PD30, and PD60, accordingly. These simulations consider the both ~~the~~ radiative and the chemical aspects of the volcanic forcing (named full aerosol effect in the following) ~~. Therefore the model was forced with~~ based on SAD values and optical properties generated by the AER model. Note ~~, that the~~ that in this study we do not include the effects of the enhanced aerosol in reducing photolysis, and related composition changes. As explained in Sect. 2.2, the PD15 ensemble simulation ~~closely resembles the conditions of~~ represents an upper limit for the effects from the Mt. Pinatubo eruption in 1991. ~~Moreover, the~~ The PI60 ensemble is comparable to the Tambora eruption in 1815 (Gao et al., 2008) although the date of the eruption and the shape of the aerosol forcings is different (Arfeuille et al., 2014). All aerosol forcing time series were applied as zonal monthly means between 690 and 3.8 hPa and interpolated to the model levels in SOCOL-MPIOM.

Furthermore, the effect of stratospheric aerosols on the atmosphere ~~will be separated into radiative perturbations (RAD~~is separated into radiative-dynamical perturbations (RAD-DYN) and changes related to heterogeneous chemical reactions ~~(HET~~on aerosol surfaces (HET-AER). In the ~~RAD~~ RAD-DYN experiment, only the optical properties of the aerosols was applied as forcing and the pre-eruption values were used for the SAD. The ~~HET~~ HET-AER experiment was forced only with time-varying SAD with the optical properties representing pre-eruptive conditions. Ensemble experiments with 8 members (Table 1) for both processes were performed for the three eruption sizes of 15, 30, and 60 Tg of SO_2.

In the following these experiments are identified by the suffix _HET-AER and _RAD-DYN. A summary of the experiments used in this study is given in Table 1.

Finally, we extract the simulated ozone changes for the full forcing and ~~RAD~~ RAD-DYN experiments and apply them as forcing in an additional set of ensemble simulations. A configuration of SOCOL-MPIOM without interactive chemistry is used in these experiments (Muthers et al., 2014b). Since ozone concentrations are not allowed to change, these simulations ~~show the pure~~ isolate only the effect of the ozone changes on the dynamical perturbation. Consequently, aerosol forcings represent pre-eruptive conditions. Ozone concentrations are applied as daily mean values on the model grid, to avoid errors due to the vertical interpolation between pressure levels and model level. These experiments were performed for the 15 and 60 Tg aerosol forcing only.

The analysis presented mainly focuses on the first winter (DJF) after the eruption. Results are always expressed in terms of anomalies to the average of the control ensemble simulations for each climate ~~state~~setting. Significance estimates are based on a two-tailed Student's ~~t test.~~ t-test using the 5 % significance level.

3 Results

3.1 Ozone changes

3.1.1 Present day conditions

The response of the global averaged column ozone (Fig. 2) reveals clear differences between the ~~radiative and the chemical effects~~RAD-DYN and the HET-AER effect. In a present day ~~climate state~~ setup (Fig. 2 top) heterogeneous chemical reactions on the aerosol surface cause a depletion of global column ozone, which is significant for more than two years. Ozone is continuously reduced for about 9 month after the eruption, independent from the eruption size. However, the amplitude of the anomaly increases with the eruption size to -13, -18, and -21 DU. The recovery phase lasts about 24 months in all simula-

tions, again independent of the eruption size. ~~The~~ As expected the spatial analysis of the ~~anomalies reveals amplified~~ HET-AER anomalies shows largest ozone depletion in the high latitudes of both hemispheres (Fig. 3a), in particular during ~~the winter months~~ spring and the polar ozone depletion through HET-AER processes increases with the eruption size (Fig. ~~3a~~S1a,d,g in the supplementary material).

Zonally averaged height profiles for the first winter after the eruption reveal that the aerosol heterogeneous chemical effect also leads to positive anomalies in the upper stratosphere above 30 hPa (Fig. 4a). However, these anomalies are present only in the first winter after the eruption and are to some extent compensated by negative anomalies in the lower stratosphere. The positive anomalies in the upper stratosphere are related to the slowdown of the NO_x cycle of catalytic ozone destruction, as N_2O_5 is converted into HNO_3 on the H_2SO_4 aerosols. By reducing the NO_x concentrations the reaction also slows down the deactivation of chlorine, which dominates ozone destruction in the lower atmosphere and explains the negative anomalies below 30 hPa. Furthermore, the conversion stops, when all N_2O_5 is consumed (Tie and Brasseur, 1995) . In the lower stratosphere N_2O_5 is quickly consumed in the months after the eruption, first in the tropical latitudes, but about 5 months after the eruptions N_2O_5 is reduced by more than 80 % at all latitudes below ~~< 30~~around 30 hPa (not shown). In the NH and SH polar stratosphere in late winter and spring the heterogeneous reactions on the aerosol surfaces and on PSCs strongly increase the chlorine concentrations in the lower stratosphere and explain the pronounced reductions ~~of~~ in ozone.

The ~~RAD~~ RAD-DYN effect causes positive global mean column ozone anomalies after the eruption, which peak about 7 months after the beginning of the eruption ($+5$, $+8$, and $+12$ DU), and return to background conditions after about 18 months (Fig. 2). Similar to the chemical effects, no clear differences in terms of the duration are found between the three eruption sizes. The column ozone anomaly time series furthermore reveal some fluctuations, mainly during the first year. In February–March and July–August, the positive column ozone anomalies undergo clear reductions. These variations are related to the present day polar ozone depletion in the NH and SH, which is further intensified after the eruption by colder conditions inside the polar vortices and chlorine activation on the PSCs ~~.~~ (Fig. 3b).

However, the polar ozone depletion in the ~~RAD~~ RAD-DYN experiment is much weaker than the signal found in the ~~HET~~ HET-AER ensemble experiment. For the 15 Tg aerosol forcing the polar ozone depletion is rather moderate (Fig. 3b), but the polar ozone depletion intensifies and lasts longer with increasing forcing strength ~~.~~ (Fig. S1e and S1h).

Overall, the spatial pattern of ozone anomalies due to the ~~RAD~~ RAD-DYN effect is more heterogeneous than for the ~~HET~~ HET-AER effect (Fig. 3b). Reduced ozone column abundances are found at tropical latitudes and increasing concentrations at mid- to high-latitudes. The tropical reduction is related to pronounced ozone depletion at 30 hPa, which is partly compensated by positive ozone anomalies above and below (Fig. 4b). This equatorial anomaly pattern is very similar for all post-eruption seasons and remains significant until the end of the first year after the eruption ~~.~~ (Fig. 3b). The circulation changes in the stratosphere that are responsible for the ozone anomalies are detected in the residual mean circulation anomaly (Andrews et al., 1987). The enhanced vertical transport of ozone changes the vertical ozone profile and replaces ozone enhanced air at 30 hPa by ozone depleted air from lower levels. Air with enriched ozone from 30 hPa further increases ozone concentrations at 10 hPa and above. The upward motion in the tropics is balanced by descending air masses in the mid-latitudes. Since these air masses originate from tropical latitudes, they transport ozone enriched air into the lower stratosphere of the mid-latitudes and create positive ozone anomalies. This meridional transport is visible in the positive anomalies of column ozone, which first occur in subtropical latitudes and reach the high latitudes several months later ~~.~~ (Fig. 3b). A fraction of descending ozone is recirculated into the lowermost tropical stratosphere and leads to positive ozone anomalies at 70 hPa. Furthermore, changes in the incoming UV radiation and photodissociation by the high optical depth of the aerosols may affect ozone production in the tropics (Pitari and Rizi, 1993), but this process is not yet implemented in the model.

The full aerosol effect displays the combined influence of both processes (Fig. 2). In the first 7 months after the eruption, ~~radiative~~ radiative-dynamical effects dominate the response of ozone with positive ozone anomalies, which reach maximum values of +2, +3, and +6 DU between autumn and early winter depending on the 15, 30, and 60 Tg forcing, respectively

(Fig. 2). In the following months, the influence of the ~~radiative~~ radiative-dynamical effects weakens, while chemical effects are still present and column ozone anomalies become negative for about two years. The imprint of the intensified polar ozone depletion is clearly visible in the full forcing experiment. With increasing eruption size the amplitude of the negative and positive anomalies increases, while the spatial patterns remain similar (Fig. 3~~e and d).~~ d and S1f,i). The dynamical perturbation of the residual circulation by the RAD-DYN effects is also clearly visible in the full forcing experiments (Fig. S3a-c).

3.1.2 Preindustrial conditions

Changing the climatic background conditions from present day to preindustrial has a strong impact on the ~~HET~~ HET-AER effect (Fig. 2 bottom and Fig. 3e). Without pronounced amounts of ODS in the stratosphere, the effect of heterogeneous chemical reactions on the chlorine cycle of ozone depletion is ~~very~~ weak. Instead, the slow-down of the NO_x cycle of ozone depletion becomes important, explaining slight positive anomalies of column ozone, for a few months after the eruption. However, with maximum anomalies between $+4$ to $+5$ DU the response is not very pronounced. The size of the eruption has no significant effect on the ozone anomalies due to HET-AER effects (Fig. 2 bottom and S2d,g).

~~RAD~~ RAD-DYN effects introduce again positive anomalies of column ozone after the eruption (Figs. 2 and 3f). In comparison to the present day ensemble simulations, the anomalies are stronger (+6, +14, +18 DU), longer lasting and the variability is lower, which is explained by the reduced polar ozone depletion in a preindustrial atmosphere. In fact, the positive anomalies of column ozone cover all latitudes from the subtropics to polar areas (Fig. 3f, compare also S2e,h,l). Consequently, the combined response is dominated by the ~~radiative~~ RAD-DYN effect and the spatial patterns of the anomaly are very similar (Fig. 3g). For larger eruption sizes, the amplitude of the ozone changes due to ~~radiative~~ RAD-DYN effects increases, while heterogeneous chemical reactions on the aerosols are only marginally affected by the eruption strength (Fig. 2).

3.2 Temperature and dynamics

3.2.1 Present day conditions

One motivation of this study is ~~the question how~~ to ask how the ozone changes described in Sect. 3.1 modulate the dynamical perturbation of the stratosphere due to the volcanic eruption.

Temperature changes associated with an ozone loss due to the ~~HET~~ HET-AER effect are small compared to the aerosol direct radiative effect, but significant temperature reductions are found in the present day ~~climate state~~ experiments for all three eruption sizes. For the 15 Tg forcing the cooling reach a minimum of -1 K in the subtropical latitudes of both hemisphere in winter (Fig. 5a). These anomalies increase to more than -2 K for the 60 Tg forcing (not shown). The reduction of the meridional temperature gradient leads to a significant slow-down of the westerly circulation in the polar stratosphere during boreal winter (Fig. 6a) and a weakening of the polar vortex. As index for the NH polar vortex intensity, time series of the zonal mean wind component at 60° N and 10 hPa (Christiansen, 2001, 2005) are shown in Fig. 7. The weakening of this ~~ū60~~ index due to the ~~HET~~ HET-AER effect is mainly a phenomena of the ~~mid-winter (December, January), while during late winter~~ mid to late winter (January, February). In January the vortex intensity reduces to 35 ± 15 m/s in the PD15_HET-AER experiment in comparison to 48 ± 11 m/s in the CTRL experiment (32 ± 18 and 36 ± 11 m/s for the 30 and 60 Tg experiment,respectively. Compare Fig. 7). During spring a slight, but not significant vortex intensification is found for the stronger forced ensemble simulations. ~~This late winter~~ In March mean value of the vortex intensity in CTRL is 9 ± 18 m/s, while the vortex in PD15_HET-AER reaches an average of 14 ± 15 m/s (21 ± 19 and 19 ± 13 m/s for the 30 and 60 Tg experiment,respectively). This spring intensification is probably related to the intensified ozone depletion and associated cooling at polar latitudes. The size of the eruption has no systematic influence on the vortex changes through the HET-AER effect. The HET-AER effect furthermore causes slight positive temperature anomalies in the NH polar stratosphere (Fig. 5a), which are related to the weakening of the polar vortex.

The ~~RAD~~ RAD-DYN effects dominate for the temperature perturbations after the eruption. Significant positive temperature anomalies are found at almost all altitudes and latitudes, with the exception of the polar areas (Fig. 5b). As expected, the temperature anomalies increase with rising aerosol mass. ~~From~~ At 50 hPa the maximum temperature anomaly, which occurs around December, is 9.5 K for the 15~~to 30~~ Tg ~~of the temperature increase is almost linear, but for the~~ and increases to 18.2 and 21.7 K for the 30 and 60 Tg ~~eruption the temperature response seems to saturate.~~ eruptions, respectively. In the troposphere, the expected cooling due to the reduced incoming short-wave flux is found at tropical and subtropical latitudes. The stratospheric warming, however, causes only a weak intensification of the NH polar vortex (Figs. 6b and 7). Significant positive anomalies are found only for a short period in late winter ~~in~~ (February, March) in the case of the 15 Tg aerosol forcing. ~~In~~ By contrast the ensemble simulations with the 30 and 60 Tg aerosol forcings ~~show~~ reveal a significant intensification of the vortex during most of the winter (Fig. 7).

The amplitude of the temperature change through the ~~HET~~ HET-AER mechanism is much weaker than the changes caused by the ~~RAD effect .~~ RAD-DYN effect (Fig. 5a). Nevertheless, the temperature reduction causes a significant weakening of the NH polar vortex, but ~~no significant increase is found in the RAD~~ only a slight increase in the vortex intensity is found for the RAD-DYN experiment for the 15 Tg aerosol forcing ~~. The different dynamic responses are~~ (Fig. 7). The difference in the response of the NH polar vortex is not yet fully understood. It may be related to the different patterns of the temperature anomaly. The aerosol induced warming covers all latitudes up to 60° N in the lower and middle stratosphere and reaches even polar latitudes in the upper stratosphere. ~~Contrary~~By contrast, the cooling associated with the ~~HET~~ HET-AER effect is limited to ~~latitudes < 30°~~the SH and up to 30° N due to the seasonal cycle of the Brewer-Dobson circulation.

The combined influence of ~~HET and RAD~~ HET-AER and RAD-DYN mechanisms is again visible in the full forcing experiment. The stratospheric warming is slightly reduced by heterogeneous chemical reactions (8.7 K in PD15) and the dynamical changes are weaker in comparison to the ~~RAD~~ RAD-DYN experiment (Fig. 6c). In particular the NH polar vortex

weakening in ~~mid-winter due to the HET~~ January and the strengthening in March caused by the HET-AER effect is clearly visible in the $\bar{u}60$ index (Fig. 7).

3.2.2 Preindustrial conditions

Under preindustrial conditions the response to an eruption differs in several aspects. Similar to the ozone anomalies, the ~~effect of heterogeneous chemical reactions~~ HET-AER effect on the stratospheric temperatures ~~and~~ (Figs. 5e and i) and dynamics is very small (Figs. ~~5e,~~ 6e, and 7). No significant anomalies are found, not even for the strongest forcing scenario ~~.~~ ~~RAD~~ although the tendency of a slight NH polar vortex weakening is apparent, in particular in late winter.

RAD-DYN effects in a ~~preindustrial~~ present day atmosphere slightly differ from the response under present day conditions. ~~The~~ At 50 hPa the maximum tropical stratospheric warming is ~~weaker in the preindustrial atmosphere~~ 1.0 K larger in the present day atmosphere (Fig. 5j) and this differences increases to 2.3 and ~~differences in the temperature anomalies increase with eruptionsize~~1.7 K for the 30 and 60 Tg eruption. The stronger warming under ~~preindustrial~~ present day conditions is not related to the dynamical ozone changes in the ~~RAD~~ RAD-DYN ensemble experiment, as will be shown below ~~.~~ (Sect. 3.3). A possible explanation could be found in the different background states. Due to the ozone depletion in the present day atmosphere, stratospheric ozone concentrations are substantially reduced in the present day tropical lower stratosphere. Furthermore, ~~the increased GHGs concentrations differ between the two climate states and both effects~~ GHGs are higher for present day conditions and both differences lead to a colder tropical stratosphere~~under present day conditions~~. With a colder stratosphere and a warmer troposphere the radiative heating from below is stronger in the present day ~~climate state.~~ atmosphere. For the combined response in a preindustrial climate ~~state~~ setting no differences to the ~~RAD~~ RAD-DYN results are found, neither for the 15 Tg (Fig. 5g) nor for the ~~60aerosol forcing.~~ stronger forcings.

~~A summary of the dynamical perturbation caused by RAD and HET and the role of the climate state and the eruption size is given in~~ In terms of the NH polar vortex, the largest

differences between preindustrial and present day conditions are found between the two CTRL experiments (Fig. **??**, which shows weakening or intensification of the meridional pressure gradient at 407). In January the vortex is slightly stronger in the present day experiment (PD_CTRL: 48±11 and the stratospheric polar low. The HET mechanism leads to a reduction of the meridional pressure gradients under present day conditions, with significant anomalies between 0 and 40m/s; PI_CTRL: 47±10 N. Differences between the eruption sizes are not found. Under preindustrial conditions, ozone changes due to heterogeneous chemical reactions have no significant influence on the meridional gradient. m/s), while during March a pronounced and significant weakening of the vortex is obvious for present day (9±18 vs. 31±16 m/s). These differences are also related to the stratospheric temperature differences between preindustrial and present day caused by different GHGs concentrations. Consequently, pronounced differences for the RAD-DYN or full forcing effect would be found between preindustrial and present day, when the vortex response is expressed in terms of anomalies to the corresponding CTRL.

The radiative effect of the aerosols increases the pressure gradient in both climate states. Geopotential height differences between low and high latitudes increase with eruption size, although not in a linear way. The anomalies are significant from the equator to the northern mid-latitudes, but not in the polar area. The weakening of the stratospheric polar low is therefore not significant, not even for the strongest eruptions size. The full aerosol effect on the meridional geopotential height gradient is in all cases very similar to the radiative effects.

3.3 Effects of the coupling between ozone and stratospheric dynamics on the stratosphere

The simulated ozone changes due to the RAD RAD-DYN effect or in the combined full forcing response could amplify or weaken the dynamical perturbation of the stratosphere. However, from the results described above, possible feedbacks between the ozone chemistry and the dynamics are difficult to extract. Two additional sensitivity ensemble experiments were therefore performed, driven only by the simulated ensemble mean ozone

changes obtained from the ~~RAD~~ RAD-DYN and full forcing experiments ~~, which were~~ (shown for instance in Figs. 2 and 3).

Zonal mean temperature anomalies (Fig. 8) reveal an amplification of the ~~aerosol-induced warming~~ RAD-DYN heating in the lower tropical stratosphere by the ozone chemistry. At these levels, ozone anomalies up to 0.2 ppmv (25 %) are found in the PD15_RAD-DYN ensemble mean, resulting in temperature anomalies of about 1.2 ± 0.3 K (80 hPa) during the first winter season after the eruption ~~.~~ (Fig. 8a). Ozone anomalies and temperature anomalies increase with eruption size and reach 0.4 ppmv and 2.9 ± 0.5 K for the 60 Tg eruption ~~in a present day climate state.~~the present day experiments (Fig. 8c). For the ozone changes extracted from the full forcing experiment, the impact on tropical stratospheric temperatures is always weaker, due to the ozone depleting effect of heterogeneous chemical reactions ~~.~~ (Fig. 8b,d). Under preindustrial conditions, the response is slightly stronger, but the differences are not significant ~~.~~ (Fig. 8e-h).

~~RAD~~ RAD-DYN ozone changes in the tropics furthermore produce a cooling above 30 hPa with temperature anomalies from -2 K (15 Tg) to -4 K (60 Tg). Ozone changes in this region therefore weaken the warming effect of the aerosols. Heterogeneous chemical reactions have no effect on temperatures in this region and ozone changes from the ~~RAD~~ RAD-DYN and the full forcing ensemble simulations lead to very similar temperature patterns. Temperature anomalies in a preindustrial atmosphere do not significantly differ from the results obtained under present day conditions. Note, that the cold anomaly in the upper stratosphere and mesosphere (present in all panels of Fig. 8) can not be attributed to volcanic induced ozone anomalies but is related to the missing diurnal cycle of ozone variations in SOCOL-MPIOM without interactive chemistry (Muthers et al., 2014b).

Furthermore, positive temperature anomalies are found in the NH polar stratosphere, though not significant except for one experiment (ozone changes from PD15_RAD-DYN). Since ozone anomalies in the NH polar stratosphere are very weak during the first winter after an eruption, these warm anomalies are probably not related to the ozone chemistry, but to dynamical changes.

The effect of the ozone perturbations on the NH polar vortex is in general small (Fig. 7) and not always consistent. Nevertheless, all experiments under present day conditions reveal a slight weakening of the vortex, which is significant for a few days in mid-winter~~. The~~ , and a slight (non significant) intensification in late winter. The size of the anomalies is roughly comparable to the changes related to the HET-AER effect. The response of the vortex to the ozone changes from the RAD-AER and full forcing experiment is weaker and not significant when ensemble simulations under preindustrial conditions are considered.

4 Discussion and conclusions

This study addresses the role of ozone changes for the dynamical perturbations of the stratosphere after strong tropical volcanic eruptions. Thereby, the underlying mechanisms and the influence of the climate ~~state~~ setting and the eruption strengths are considered. The results are based on a number of ensemble sensitivity simulations with the AOCCM SOCOL-MPIOM, which allows us to separate the effect of heterogeneous chemical reactions from the warming effect of the aerosols for ~~two different climate states and~~ preindustrial and present day conditions as well as three different eruption intensities.

In agreement with earlier studies we find that both processes, heterogeneous chemical reactions on the aerosol surface and the warming effect of the aerosols are important for the ozone changes after the eruption in an atmosphere with enhanced concentrations of ODS (Pitari and Rizi, 1993; Al-Saadi et al., 2001). ~~The chemical effect, however, is the dominant one with a pronounced global reduction of the total column ozone after the eruption, when the atmosphere contains high loads of ODS. Regionally, the warming effect can become more important, for instance~~ A comparable case-study for the 1991 Mt. Pinatubo was performed by Aquila et al. (2013) , with a similar separation between radiative-dynamical and heterogeneous chemical effects on the aerosols. They identified a combination of HET-AER and RAD-DYN processes to be responsible for the ozone anomalies in the SH. In particular they found similar anomalies in the residual mean circulation being responsible for reduced ozone in the tropics and ~~mid latitudes, where dynamical changes cause~~

pronounced negative (equator) and positive (subtropics)anomalies of column ozone. Under enhanced ozone concentrations at mid-latitudes (compare Fig. 4b and their Fig. 7). This response, however, is limited to the early phase of the eruptions and in combination with the phase of the Brewer-Dobson circulation the anomaly-pattern is found only in the SH. In boreal winter, when tropical stratospheric temperature anomalies are at maximum in our experiments, our findings suggest a similar response of the residual mean circulation in the NH with positive column ozone anomalies at mid-latitudes. Aquila et al. (2013) did not simulate positive column ozone anomalies in the NH related to radiative-dynamical changes (compare their Fig. 6) and concluded that NH ozone anomalies are mainly affected by HET-AER effects. This difference in the response is not yet understood, but may be related to differences in the aerosol forcings. Understanding the response to the RAD-DYN mechanism is of particular importance for volcanic eruptions under preindustrial conditions with low load of ozone depleting halogens, the where chemical effects become very weak and the response is dominated by the aerosol warmingradiative-dynamical effects.

The dynamical perturbation of the stratosphere is dominated by the radiative effects RAD-DYN effect of the aerosols. During winter time, the warming in the tropical stratosphere increases the meridional temperature gradient towards the poles and strengthens the westerly circulation in the polar stratosphere. Ozone changes, related to heterogeneous chemical reactions on aerosols under present day climatic conditions have been shown to conditions weaken the warming in the tropical stratosphere and the NH polar vortex. ContraryBy contrast, ozone changes related to the RAD RAD-DYN mechanism amplify the warming in the subtropical and mid-latitude lower stratosphere, but weaken the temperature response in the middle stratosphere. The latter causes a slight weakening of the polar vortex. In agreement with Stenchikov et al. (2002); Shindell et al. (2003) we found an intensification of the NH polar vortex in late winter due to ozone changes after strong volcanic eruptions in a present day atmosphere. Our results show that both, HET-AER and RAD-DYN effects contribute to this intensification.

The climate state atmospheric composition influences the response of the ozone chemistry and the dynamical response to the volcanic aerosols in a significant way. As already

shown by Tie and Brasseur (1995) heterogeneous chemical reactions on the aerosol surface cause pronounced global ozone depletion when stratospheric loads of ozone depleting halogens are high, but has only a weak positive effect in a preindustrial atmosphere. Furthermore, ozone depletion scales with the amplitude of eruptions ~~in a present day climate state~~under present day conditions, but for ~~preindustrial conditions~~ a preindustrial atmosphere the response is independent from the eruption size (Tie and Brasseur, 1995). With the projected reduction of ODS (Austin and Wilson, 2006; Eyring et al., 2007; Austin et al., 2010) the effect of heterogeneous chemical reactions can be expected to become less important for future eruptions. However, differences in the background state affect the results of this study as well. In recent decades, the stratosphere has undergone a pronounced cooling trend (e.g., Thompson et al., 2012), related to the ozone changes and the increasing levels of GHG (Shine et al., 2003). This cooling also affects the meridional temperature profiles in the stratosphere and the dynamics. In this study, significant differences in the vortex behaviour exist between the control simulations for preindustrial and present day, and these differences ~~are often larger ,~~ can be larger than the differences between the simulations perturbed by the volcanic eruption.

A direct comparison of our results to observations is difficult given the highly idealised character of our experiments. Nevertheless, one experiment, the 15 Tg experiment for the full aerosol effects ~~in a present day climate state, is characterised by very similar boundary conditions as the eruption of~~ under present day conditions, can be considered similar to the perturbation from Pinatubo in 1991. Globally, the combined aerosol effect for the 15 Tg aerosol forcing results in an ozone loss of 10.4 ± 3.7 DU 21 months after the start of the eruption. This agrees reasonably well with observations for Pinatubo (Bodeker et al., 2005), where a reduction of -13.9 DU was observed peaking in May 1993 (22 months after the eruption). Furthermore, the pattern of column ozone anomalies in Bodeker et al. (2005) is similar to the pattern found in the PD15 ensemble mean (Fig. 3). After the eruption, negative anomalies are found in the tropics and positive anomalies in the sub-tropics and mid latitudes. The magnitude of both is stronger in SOCOL-MPIOM than in observations. In particular, the positive column ozone anomaly in the NH mid-latitudes is weaker in the Bodeker

et al. (2005) data set. This may be related to the QBO effect (Telford et al., 2009; Randel and Wu, 1995) or the Brewer–Dobson circulation (Aquila et al., 2013), which modulated the ozone anomalies after Pinatubo. The modulating effect of the QBO, however, has been removed by the comparison to an ensemble of control simulations nudged by the same QBO reconstruction. Overestimated ozone anomalies in the PD15 ensemble may further be related to the too strong warming simulated in the tropical stratosphere by SOCOL-MPIOM. The remaining anomalies, i.e., the polar ozone depletion and the global reduction of columns ozone about one year after the eruptions agrees reasonable well with the simulated anomalies. For the vertical pattern of ozone changes (e.g., Hassler et al., 2008) the agreement between model results and observations is also reasonable, with the exception of stronger anomalies in the model, in particular in the tropics.

A caveat of this study is the overestimated warming in the tropical stratosphere, which may ~~leads~~ lead to an overestimation of the dynamical response and the resulting ozone changes. This needs to be considered ~~,~~ when interpreting the results of the ~~RAD~~ RAD-DYN and full forcing ensemble results. An overestimation of the lower stratospheric warming after volcanic eruptions is a feature common to many models (e.g. Driscoll et al., 2012). The maximum warming after the Pinatubo eruption in a multi-model ensemble average of 13 CMIP5 models (model selection as in Driscoll et al., 2012) is 3.8 ± 2.9 K for the temperature anomaly at 50 hPa, while reanalysis products and observations suggest temperature anomalies between 2.5 and 3 K (Labitzke and McCormick, 1992; Dee et al., 2011). However, the phase of the QBO and other dynamical processes are suggested to reduce the stratospheric warming after Pinatubo (Driscoll et al., 2012; Fueglistaler et al., 2014). SOCOL-MPIOM simulates a warming of 8.7 ± 1.2 K in the PD15 ensemble. Our results show that the overestimated warming is to some extent related to the ozone changes, which amplify the warming by about ~~1.5~~0.9 K (15 Tg forcing ~~). Furthermore, the~~ at 50 hPa). The aerosol forcing applied in this study also contributes to the overestimated warming. We use ~~an~~ a 2D global aerosol forcing produced by an aerosol micro-physical model, which allows us to generate physical consistent forcing for different eruption intensities. This model-generated forcing differs from satellite based observations for Pinatubo in several aspects (as described by Arfeuille et al.,

2013). For comparison, we also perform eight simulations forced by realistic satellite based aerosol concentrations for the Mt. Pinatubo eruption (SAGE_4λ in Arfeuille et al., 2013). In these simulations (not shown) the stratospheric warming is still overestimated (maximum temperature anomalies: 6.0 ± 1.0 K), but the agreement to ~~other~~ CMIP5 is better, in particular when the temperature effect of the ozone changes is considered.

In summary, we show that ozone ~~changes after strong tropical volcanic eruptions play a minor role for the dynamical perturbation of the stratosphere although for in same cases (e. g.~~ is affected globally by a volcanic eruption for several years. Both effects, the radiative dynamical perturbation by the volcanic aerosols as well as heterogeneous chemical reaction on the aerosols are important for the response of the ozone chemistry. The climate setting, in particular the atmospheric concentrations of ODS, has the strongest effects on the heterogeneous chemical effect on aerosol surfaces with pronounced global ozone depletion for present day ODS concentrations (peak reductions of -13, ~~high load of ODS)a weak influence is detectable. Nevertheless, this influence is small compared~~ -18, and -21 DU for the 15, 30, and 60 Tg eruptions, respectively) and slight ozone increase for preindustrial ODS concentrations (between 4–5 DU for all eruptions). Radiative dynamical ozone changes are positive for preindustrial and present day conditions, but for present day the response is weakened by amplified polar ozone depletion (+5,+8,+12 DU peak column ozone anomalies for present day and +6, +14, +18 DU for preindustrial). The full effect of the volcanic aerosol, therefore, clearly differs between preindustrial and present day, with long lasting ozone depletion in a present day atmosphere and positive ozone anomalies for preindustrial conditions. The response of stratospheric temperature and dynamics is dominated by the radiative heating effect of the aerosols. A small influence of the climate setting on the heating of the lower tropical stratosphere was found, with larger temperature anomalies for the present day experiments. Dynamical radiative ozone changes further amplify the stratospheric temperature anomalies in the lower tropical stratosphere (and cause a cooling in higher levels). Ozone changes due to heterogeneous chemical reactions on the aerosols are responsible for a slight cooling of the tropical stratosphere. In winter and early spring after the eruption, the NH polar vortex is intensified, due to the radiative

~~effect of the aerosols.~~ warming in the tropical stratosphere. Ozone changes, either due to radiative-dynamical effects or heterogeneous reaction on the aerosol surface, induce a slight weakening of the vortex in mid-winter. In late winter they cause a slight strengthening of the westerly circulation in the NH polar stratosphere.

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Table 1. Overview of the ensemble experiments used in this study. SO_2 mass refers to the aerosol amount assumed in the generation of the volcanic forcing (compare Sect. 2.2). Climate ~~state~~setting: PD: 1990s conditions with high concentrations of ozone depleting substances (ODS). PI: preindustrial atmosphere with low concentrations of ODS. Volcanic forcing: SAD: surface area density of the aerosols. OP: optical properties, i.e., extinction rates for all spectral intervals. For each ensemble, 8 simulations are performed.

Ensemble	SO_2 mass [Tg]	Climate ~~state~~ setting	Volcanic forcing	Atm. chemistry
PD[15,30,60]	15, 30, 60	PD	SAD and OP	interactive
PI[15,30,60]	15, 30, 60	PI	SAD and OP	interactive
PD[15,30,60]_HET-AER	15, 30, 60	PD	SAD	interactive
PI[15,30,60]_HET-AER	15, 30, 60	PI	SAD	interactive
PD[15,30,60]_RAD-DYN	15, 30, 60	PD	OP	interactive
PI[15,30,60]_RAD-DYN	15, 30, 60	PI	OP	interactive
PD[15,60]_o3	15, 60	PD	–	ozone from PD[15,60]
PI[15,60]_o3	15, 60	PI	–	ozone from PI[15,60]
PD[15,60]~~RAD~~RAD-DYN_o3	15, 60	PD	–	ozone from PD[15,60]_RAD-DYN
PI[15,60]~~RAD~~RAD-DYN_o3	15, 60	PI	–	ozone from PI[15,60]_RAD-DYN
PD_CTRL	–	PD	–	interactive
PI_CTRL	–	PI	–	interactive

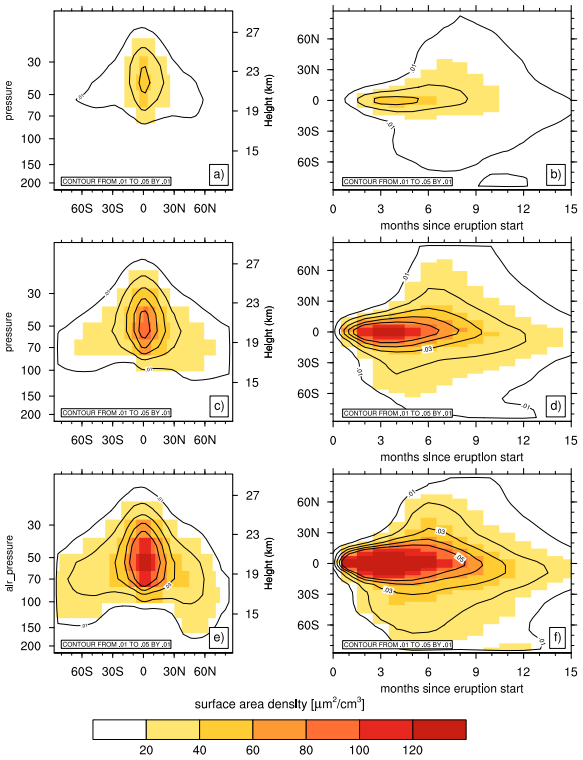

Figure 1. Aerosol data sets used in the simulations for the 15 Tg **(a, b)**, 30 Tg **(c, d)**, and 60 Tg **(e, f)** SO_2 aerosol forcing. Surface area densities (SAD) are displayed by colours; contours denote extinction rates in the visible (440–690 nm) with contours from 0 to 0.05 by an interval of 0.01 km^{-1}. The column on the left displays averages for the first post-eruption winter season (DJF) as a function of pressure and latitude, while the right column shows Hovmöller diagrams of the monthly mean SAD forcing at 50 hPa.

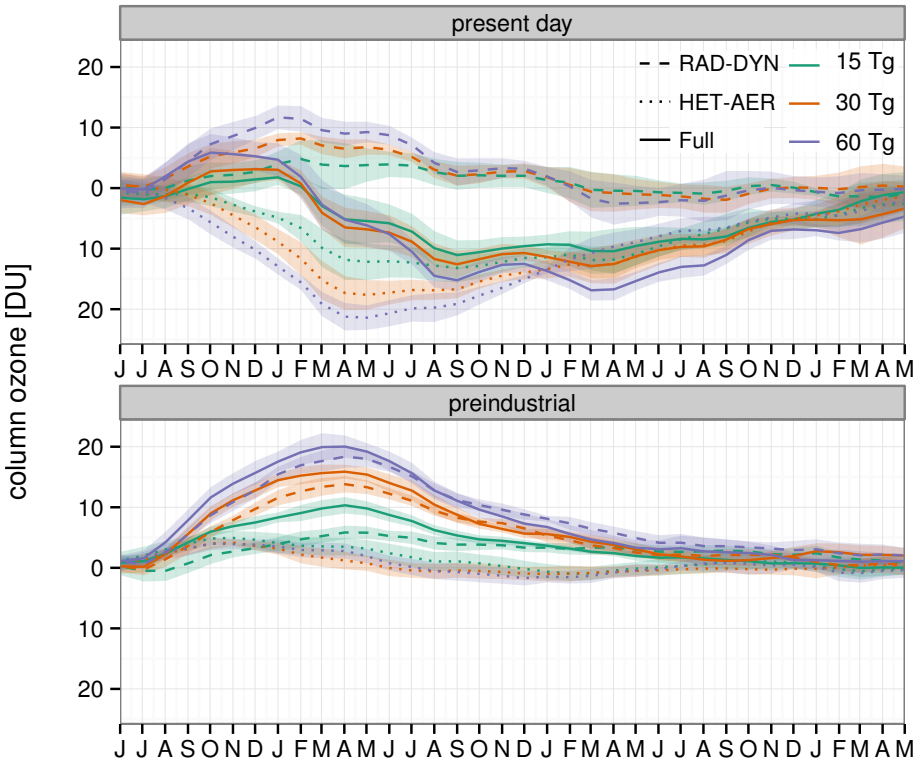

Figure 2. Monthly mean global mean anomalies of column ozone [DU] ~~in a~~ for present day (top) and preindustrial ~~climate state~~ conditions (bottom) for the ensemble simulations with the full forcing effect (Full, solid lines), the ensemble simulations considering only the heterogeneous aerosol chemical effect of the aerosols (~~HET~~HET-AER, dotted lines), and the simulations forced only by the ~~radiative~~ radiative-dynamical aerosol effects (~~RAD~~RAD-DYN, dashed lines). The different eruption sizes are indicated by colours. Lines denote the ensemble mean, shading the standard deviation ~~(SD)~~ of the ensemble.

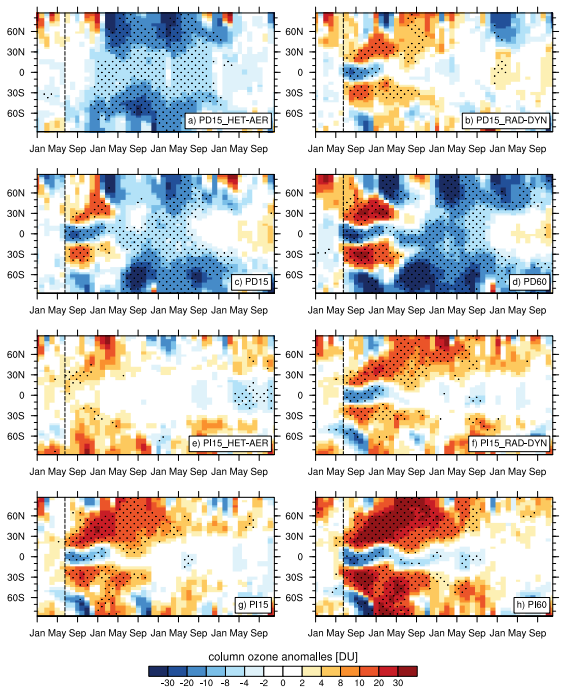

Figure 3. Zonal average monthly mean anomalies of total ~~columns~~ column ozone ~~DUbetween January of the eruption year (year 0) and 40 months after the eruption~~ in different ensemble simulations: **(a)** anomalies due to the effect of heterogeneous chemical reactions for the 15 Tg eruption ~~in a~~ for present day ~~climate state~~conditions. **(b)** Same as **(a)**, but for the ~~radiative~~ radiative-dynamical effects of the aerosols. **(c)** Same as **(a)**, but for the full forcing simulations. **(d)** Full forcing simulations for the 60 Tg aerosol forcing data set. **(e–h)** Similar to **(a–d)**, but for ~~the~~ preindustrial ~~climate state~~conditions. ~~Anomalies are calculated relatively to the corresponding control ensemble mean and the stippling~~ Stippling in the simulation panels indicates significant differences to the control (Student's t test $p \leq 0.05$). The beginning of the eruption is depicted by the vertical dashed line.

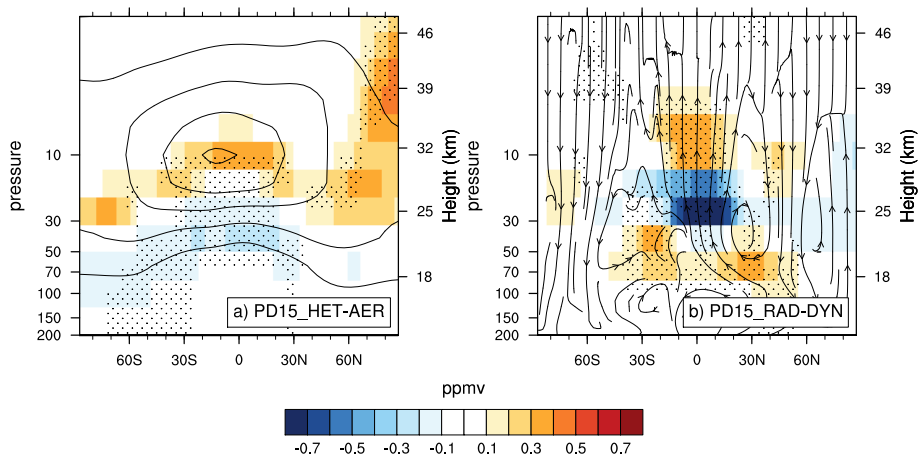

Figure 4. Ensemble mean zonal mean ozone mixing ratio anomalies [ppmv] for the first post eruption DJF season in the present day ensemble experiment considering **(a)** the effect of aerosol heterogeneous chemical reactions **(b)** the ~~radiative~~ radiative-dynamic effect with the 15 Tg aerosol forcing under present day conditions. Contour lines in **(a)** denote the climatological average DJF ozone mixing ratios in the present day control ensemble. The streamlines in **(b)** show the residual circulation anomalies (Andrews et al., 1987). Stippling indicates significant anomalies (Student's t test $p \leq 0.05$).

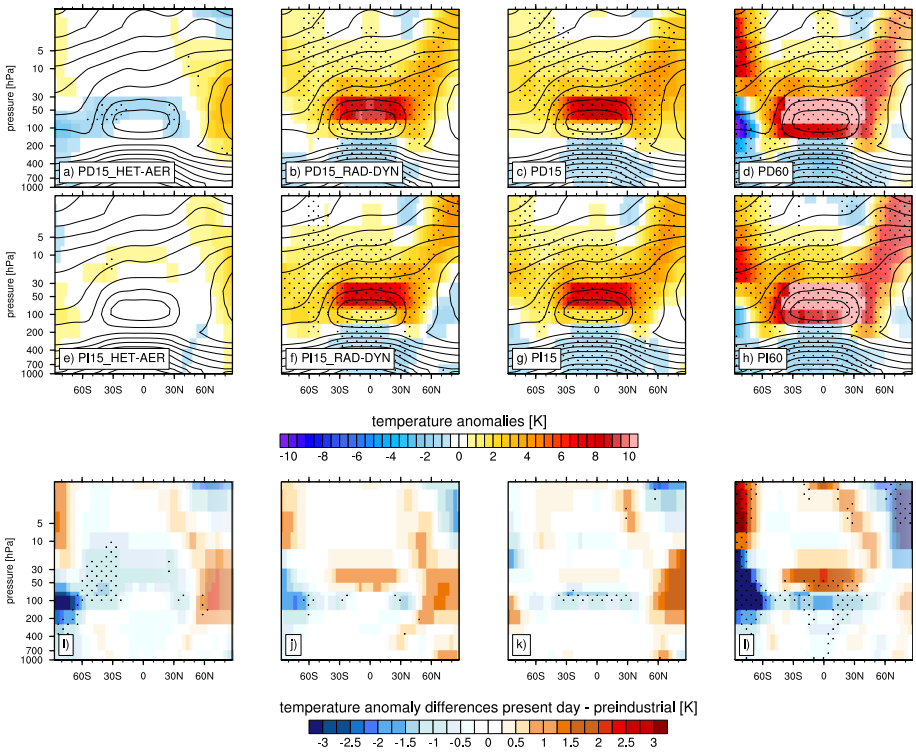
a) PD15_HET-AER
b) PD15_RAD-DYN
c) PD15
d) PD60
e) PI15_HET-AER
f) PI15_RAD-DYN
g) PI15
h) PI60
i)
j)
k)
l)
pressure [hPa]
5
10
30
50
100
200
400
700
1000
60S 30S 0 30N 60N
temperature anomalies [K]
-10 -8 -6 -4 -2 0 2 4 6 8 10
temperature anomaly differences present day - preindustrial [K]
-3 -2.5 -2 -1.5 -1 -0.5 0 0.5 1 1.5 2 2.5 3

Figure 5. Zonal mean temperature anomalies [K] for the first post eruption winter (DJF) relative to the average of the present day or preindustrial control simulations: **(a)** anomalies due to the effect of heterogeneous aerosol chemical reactions (~~HET~~HET-AER) for the 15 Tg eruption ~~in a~~ under present day ~~climate state~~conditions, **(b)** same as **(a)**, but for the radiative effects of the aerosols, **(c)** same as **(a)**, but showing the ensemble mean of the full forcing simulations, **(d)** full forcing simulations for the 60 Tg aerosol forcing data set, and **(e–h)** similar to **(a)**, but for ~~the~~ preindustrial ~~climate state~~conditions. Anomalies are calculated relatively to the corresponding control ensemble mean and the stippling in the simulation panels indicates significant differences to the control (student's t test $p \leq 0.05$). Contours denote the climatological mean DJF temperatures in the control ensembles. **(i-l)** Differences between the present day and preindustrial temperature anomalies for the different experiments.

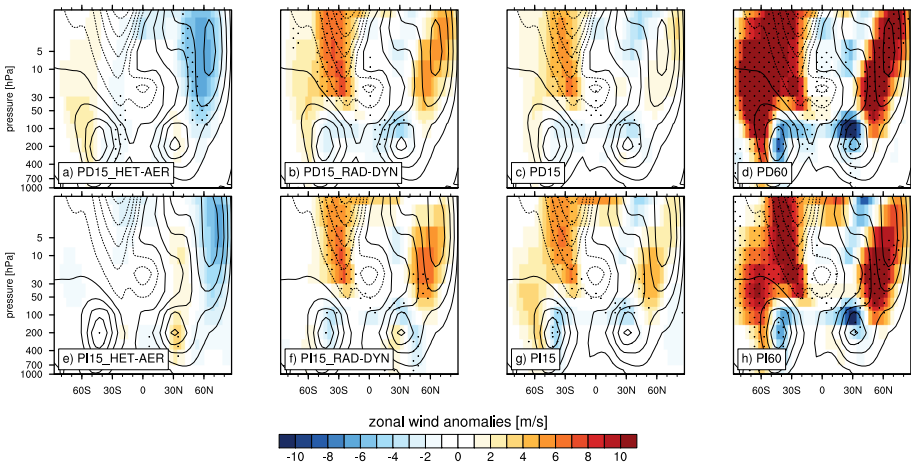

Figure 6. Similar to Fig. 5, but for the zonal mean zonal wind anomalies [$\mathrm{m\,s^{-1}}$].

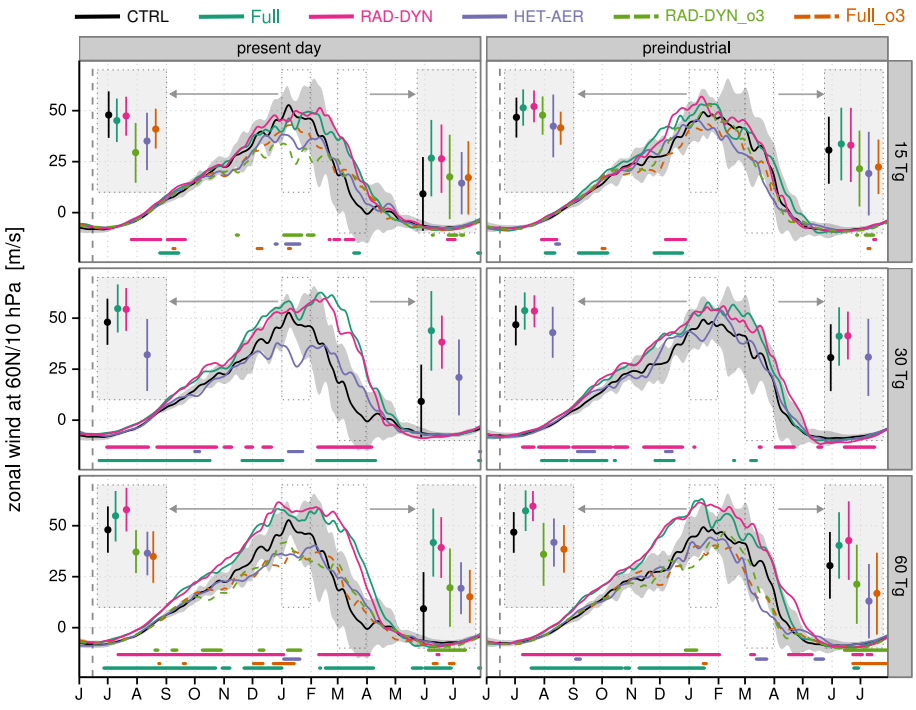
CTRL
Full
RAD-DYN
HET-AER
RAD-DYN_o3
Full_o3
present day
preindustrial
15 Tg
30 Tg
60 Tg
zonal wind at 60N/10 hPa [m/s]
50
25
0
J J A S O N D J F M A M J J
J J A S O N D J F M A M J J

Figure 7. Daily zonal mean zonal wind at 60° N [m s^{-1}] used as index for the NH polar vortex intensity. Dots at the bottom of each panel indicate days with significant differences to the control ensemble ($p \leq 0.05$). All values are smoothed with an 11 day low pass filter. The start of the eruption (mid of June) is indicated by the vertical dashed line. The shading indicates the ~~SD~~ standard deviation in the control experiments. Additionally, each panel displays the monthly mean statistics for January (left) and March (right) with the dots representing the mean value and the line range corresponding to the standard deviation for each ensemble experiment.

~~Zonal mean geopotential height anomalies mat 40for the first post eruption DJF season relative to the average of the present day or preindustrial control simulations. The grey shading indicates the SD in the control experiments. Dots at the bottom of each panel indicate significant differences to the corresponding control ensemble ($p \leq 0.05$).~~

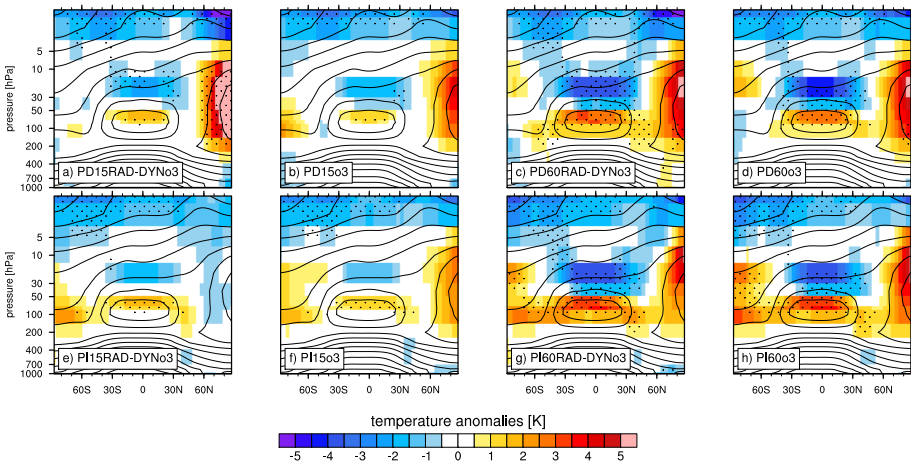

Figure 8. Similar to Fig. 5 but showing DJF zonal mean temperature anomalies [K] in the ozone sensitivity ensemble experiment. (top) present day ensemble experiments forced by ozone changes from PD15_RAD-DYN **(a)**, PD15 **(b)**, PD60_RAD-DYN **(c)**, and PD60 **(d)**. (bottom) Same forcings but for the preindustrial ~~climate state~~ experiments. Note the different colour scaling in comparison to Fig. 5.