

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 precise and we adopted your 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 instances 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 surface (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 clarified 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 whereby 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 surprisingly 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 radiatively dynamical 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 chlorine activation on the PSCs under present day conditions. As visible, for instance, in Fig. 2 the RAD-DYN timeseries differ between pre-industrial 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 replace ‘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 'tropospheric 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 occurrence and types of polar stratospheric clouds (PSCs).'

Line 22 for point iv) insert 'composition' before 'changes induced' to distinguish this from the temperature changes 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 NO_x 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 Cl_x and HO_x 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 Cl_x and HO_x 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

strengthening 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 the end of the previous 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 Rizzi, 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 varies more or less linearly with the SO₂ mass injected. I don't think that is the case. Since the warming is associated 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 sentence 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 'radiatively-dynamical 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 misleading. 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 supri- singly 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 chemical effect is not considered in these 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 can 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.
- Dhomse, S. S., M. P. Chipperfield, W. Feng, R. Hossaini, G. W. Mann and M. L. Santee (2005) Revisiting the hemispheric asymmetry in midlatitude ozone changes following the Mount Pinatubo eruption: A 3-D model study, *Geophys. Res. Lett.*, 42, 3038–3047, doi:10.1002/2015GL063052.
- 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.