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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The impact of volcanic aerosols on stratospheric ozone and the Northern Hemisphere polar vortex: separating radiative from chemical effects under different climate conditions" *by* S. Muthers et al.

Anonymous Referee #2

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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.

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.

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."

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.

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 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).

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. 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.

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."

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)."

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."

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."

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."

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 radiative-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 strengthening of the Northern Hemisphere (NH) polar vortex during the first post-eruption winter."

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

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.

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.

9) Introduction – pg 14277, lines 23-25 – sentence improved by deleting "volcanic", ",

which are", "gases" and "can" – more succinct and easier to read.

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".

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

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

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".

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".

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

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.

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".

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".

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?)."

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.

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).

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

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...".

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

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".

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."

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..."

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."

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

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

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

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)."

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

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.

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

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

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.

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.

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".

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

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

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)?

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

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".

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"

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

47) section 2.3 pg 14286 line 6 – replace "radiative perturbations (RAD)" with "radiative-dynamical perturbations (RAD-DYN)" and replace "reactions (HET)" with "reactions on

aerosol surfaces (HET-AER)".

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

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

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

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

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

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".

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

71) section 3.2 pg 14290 line 11 replace "<" with ">". Also consider adding "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....

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.

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 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?

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

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

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

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

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

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"

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

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

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

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

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

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

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?

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.

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...".

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

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

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 inmidlatitude 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.

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