

Web links to the author's journal account have been redacted from the decision letters as indicated to maintain confidentiality.

21st Dec 20

Dear Dr Osipov,

Your manuscript titled "Toba supervolcano eruption caused tropical ozone hole" has now been seen by 2 reviewers, and I include their comments at the end of this message. They find your work of interest, but some important points are raised. We are interested in the possibility of publishing your study in *Communications Earth & Environment*, but would like to consider your responses to these concerns and assess a revised manuscript before we make a final decision on publication.

In particular, both reviewers suggest that the text requires re-framing of the novel claims and further explanation in places. Please ensure that in the revised manuscript you:

- Ensure the specific claims made in your manuscript are placed firmly in the context of existing relevant literature - as highlighted by Reviewer 1 - and revise them accordingly if necessary.
- Provide further discussion of the shortcomings of the volcanic winter scenario, which would help to emphasise the novel contribution of your proposed model.
- Provide further discussion of the role of halogens and the implications for your model calculations, which exclude them. This is mentioned briefly in the submitted manuscript, but requires further detail.
- Ensure that sufficient additional explanation is added to bridge the gap between your modelled ozone losses and the inferred UV-B enhancement, as highlighted by Reviewer 2.

Although reviewers consider that the scientific basis of your manuscript is generally sound, Reviewer 2 raises specific concerns about Figure 5 which I would like to see addressed. Comparison to physical measurements made post-Pinatubo has also been suggested as a useful validation of your proposed mechanism. Although not the main subject of your modelling, some discussion of whether these measurements support your proposed mechanism would be welcomed.

We therefore invite you to revise and resubmit your manuscript, along with a point-by-point response that takes into account the points raised. Please highlight all changes in the manuscript text file.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

Please use the following link to submit your revised manuscript, point-by-point response to the referees' comments (which should be in a separate document to any cover letter) and the

completed checklist:

[link redacted]

****** This url links to your confidential home page and associated information about manuscripts you may have submitted or be reviewing for us. If you wish to forward this email to co-authors, please delete the link to your homepage first ******

We hope to receive your revised paper within six working weeks (we understand that this is coming to you ahead of the Christmas vacation period); please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we may close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

We understand that due to the current global situation, the time required for revision may be longer than usual. We would appreciate it if you could keep us informed about an estimated timescale for re-submission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to accommodate necessary extensions nevertheless.

Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Emma Liu, PhD
Communications Earth & Environment
orcid.org/0000-0003-1749-9285

EDITORIAL POLICIES AND FORMATTING

We ask that you ensure your manuscript complies with our editorial policies. Please ensure that the following formatting requirements are met, and any checklist relevant to your research is completed and uploaded as a Related Manuscript file type with the revised article.

Editorial Policy: Policy requirements

Furthermore, please align your manuscript with our format requirements, which are summarized on the following checklist:

Communications Earth & Environment formatting checklist

and also in our style and formatting guide Communications Earth & Environment formatting guide .

******* DATA: Communications Earth & Environment endorses the principles of the Enabling FAIR data

project (<http://www.copdess.org/enabling-fair-data-project/>). We ask authors to make the data that support their conclusions available in permanent, publically accessible data repositories. (Please contact the editor if you are unable to make your data available).

All Communications Earth & Environment manuscripts must include a section titled "Data Availability" at the end of the Methods section or main text (if no Methods). More information on this policy, is available at <http://www.nature.com/authors/policies/data/data-availability-statements-data-citations.pdf>.

In particular, the Data availability statement should include:

- Unique identifiers (such as DOIs and hyperlinks for datasets in public repositories)
- Accession codes where appropriate
- If applicable, a statement regarding data available with restrictions
- If a dataset has a Digital Object Identifier (DOI) as its unique identifier, we strongly encourage including this in the Reference list and citing the dataset in the Data Availability Statement.

DATA SOURCES: All new data associated with the paper should be placed in a persistent repository where they can be freely and enduringly accessed. We recommend submitting the data to discipline-specific, community-recognized repositories, where possible and a list of recommended repositories is provided at <http://www.nature.com/sdata/policies/repositories>.

If a community resource is unavailable, data can be submitted to generalist repositories such as [figshare](https://figshare.com/) or [Dryad Digital Repository](http://datadryad.org/). Please provide a unique identifier for the data (for example a DOI or a permanent URL) in the data availability statement, if possible. If the repository does not provide identifiers, we encourage authors to supply the search terms that will return the data. For data that have been obtained from publically available sources, please provide a URL and the specific data product name in the data availability statement. Data with a DOI should be further cited in the methods reference section.

Please refer to our data policies at <http://www.nature.com/authors/policies/availability.html>.

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

This is an interesting paper challenging the conventional idea of massive volcanic eruptions impacting life on Earth through the so-called volcanic winter. The authors point out the shortcomings of such a scenario. Using simulations with a state-of-the-art global chemistry-aerosol-climate model, the authors show that the Toba eruption should have led to severe stratospheric ozone losses in tropics and hence extensive increases in solar UV levels at the surface. This would have resulted in serious stresses for living cells with environmental and ecological consequences.

Studies on the impact of future large eruptions on societies and ecosystems are currently focused on the volcanic winter scenario. The results show that they should also consider an ozone-UV scenario. This kind of papers challenging conventions and paradigms fits well in Nature. However, the text is a bit missing the novel point of the paper. For instance, the title is about Toba causing ozone losses. They claimed to have discovered a “previously unaccounted mechanism that influences ozone” (also pointed out in the abstract: “hitherto overlooked mechanism”). This ozone loss mechanism is not new, only the consequences for UV levels and the emerging of a volcanic ozone-UV scenario are new. I have to recommend publication but with major revisions. I am happy with the scientific content but there is still a lot of work to do on the text, i.e. correcting misleading claims and stressing the novel scenario.

Major points:

- The authors should tone down their claim of a discovery about a “previously unaccounted mechanism that influences ozone” (also pointed out in the abstract: “hitherto overlooked mechanism”). This mechanism has been accounted before. They explain this mechanism in the text (L112-L125) and abstract: “But massive tropical volcanic eruptions such as that of Toba give rise to large amounts of SO₂ and sulfate aerosols in the stratosphere, which strongly enhance the UV extinction optical depth. The SO₂ cloud strongly absorbs UV radiation, and weakly absorbing sulfate additionally scatters the photons back to space, which decreases the radiation flux needed to form ozone within and below the volcanic plume.... Showing shutdown of the O₂ and strong reductions of the O₃ photolysis within the volcanic plume”. Bekki et al., Nature, 1993, decompose exactly this effect of SO₂ absorption of UV on O₂ photolysis, ozone production and resulting ozone losses for the Pinatubo case and Bekki, 1995 and Bekki et al., GRL, 1996, takes explicitly into account this mechanism of ozone losses for the Toba case. Michelangeli et al., JGR, 1989, takes into account the UV scattering by volcanic stratospheric aerosols for the Pinatubo case.
- The authors could discuss a bit more the alternative ozone-UV scenario and possible gains on shortcomings of the volcanic winter scenario. In particular, they should stress more that the ozone-UV scenario is not limited to the effects of volcanic sulphur. The scenario also holds for volcanic halogens with several model calculations also showing massive ozone losses with volcanic halogens.

Minor points:

- L67: “While the “missing” sulfur source remains to be found to resolve this paradox”. The “excess” sulphur paradox is not a complete mystery. We know it is probably due to the presence of an exsolved vapor phase in the magma before eruption (see 23 and Wallace, 2003] but this process is poorly understood and quantified.
- L121-122: missing word: “...showing shutdown of the O₂”, add photolysis
- L131-132 and also later: The fact that ozone anomaly was peaking and massive in the tropics was already shown in Bekki et al, GRL, 1996, in the same case (Toba).
- L187-191: Very important point that these simulations are lower limits regarding ozone losses. Volcanic halogens have been largely neglected in chemistry-climate simulations. Model simulations converge on the effect of volcanic halogens on the ozone layer with massive ozone losses: Brenna, 2019, but also Klobas, 2017, and Cadoux, 2015. Therefore,

the alternative ozone-UV scenario is not limited to volcanic sulphur but should also include the halogen component.

- The authors do not discuss stratospheric H₂O. First, it could be a limited factor in sulphur oxidation to sulphate. At the same time, the warming of the tropical tropopause (at least in models) leads to much more water entering the stratosphere and hence more ozone destruction by the every effective Hox cycles in the lower stratosphere where most of the ozone is found. What are the changes in stratospheric H₂O in their model simulations?

Reviewer #2 (Remarks to the Author):

Key results: The authors have quantified highly significant reductions in total ozone they believe accompanied the Toba eruption. Leaving aside their speculations regarding the effect of the eruption on the survivable of humanity, the major significance of this work is the increase in UV-B that might occur should a Toba-class eruption occur today.

Validity: The paper flaws that must be reasonably resolved. For example, the paper proposes both a sharp increase in UV-B resulting from major ozone destruction by Toba's emissions and their byproducts and significant blockage of UV-B by the volcanic plume. How the latter allows for the former is not clearly explained. Post-Pinatubo ozone, UV-B and AOD data are available. For example, see Fig. 5.5 at

https://www.researchgate.net/publication/227860247_Surface_ultraviolet_radiation_Past_present_and_future/figures?lo=1 Is there not a role for actual measurements such as these in conjunction with model predictions? (This link is for 40N. Dozens of UVB Biometers and Dobson spectrophotometers were in use at many locations when Pinatubo erupted.) At minimum, physical data could provide a test of model performance.

Originality and significance: The Toba eruption and its immense consequences have been reported in many papers. These papers do not always agree, and an umbrella statement to this fact might be appropriate, especially for those new to paleovolcanology. While some of these papers rely on studies and measurements of physical ejecta, others rely on model predictions and comparisons with more recent major eruptions (e.g., Tambora, Krakatau, Pinatubo, etc.). This paper falls in the latter category, with modeling forming its key analysis mechanism. I am unaware of prior work that gives claims for the magnitude of O₃ destruction and UV increase as specific as this paper. Assuming that the paper can be revised to accommodate concerns given below, their predictions should be considered by both atmospheric scientists and those who study the impact of UV-B on human health and the survivability of plant life.

Data & methodology: Figure 5 is both mistaken and troubling. (A) It is mistaken because it ascribes a loss of ozone following Pinatubo of only 5 DU. This is incorrect, for various papers have described much larger losses. For example, Grant, W. B., et al. (1994, Aerosol-associated changes in tropical stratospheric ozone following the eruption of Mount Pinatubo, J. Geophys. Res., 99, 8197– 8211) reported O₃ losses of as much as 33%. (This reviewer measured 60-day average differences at 29.6N of pre-and post-Pinatubo spring O₃ of 26 DU (-14.8%) and winter O₃ of 32 DU (-17.8%). Paper submitted.) (B) Figure 5 is troubling, for no reference or citation is provided for the 5 DU assertion

for Pinatubo. (Line 359 suggests that Fig. 5 is from the model. That should be stated in the caption unless the model is cited in the caption for Fig. 1 as suggested herein.) If the predicted O3 decreases for putative eruptions greater than Pinatubo are a product of the model(s) employed by the authors, Fig. 5 could raise questions about other results in the paper.

Validity of the approach, quality of the data and quality of presentation: Concerns about Fig. 5 are given above. Another concern is the need for more discussion to better explain the increase in UV-B that will accompany their predicted O3 losses. The paper observes that UV-B that leaks through what remains of the ozone layer will be modulated both by the attenuation of the volcanic aerosol cloud and absorption by both O3 and SO2. But how the UV-B managed to achieve ultra-high levels in the presence of the post SO2 sulfate cloud was not fully explained.

Appropriate use of statistics and treatment of uncertainties: The modeled charts in Figs. 1-4 do not include error bars, which is customary for visualizations. As for the model, the 2006 version of the GISS Model E is used in this paper. This model has been updated to GISS Model E2.2, a 2020 version that specifically advances results for elevations in the upper troposphere and stratosphere that are most impacted by Plinian volcano eruptions. The authors might want to look into this updated version of the model they used. Details are here: Rind, D., Orbe, C., Jonas, J., Nazarenko, L., Zhou, T., Kelley, M., et al. (2020). GISS Model E2.2: A climate model optimized for the middle atmosphere—Model structure, climatology, variability, and climate sensitivity. *Journal of Geophysical Research: Atmospheres*, 125, e2019JD032204. <https://doi.org/10.1029/2019JD032204>

Conclusions: Please see suggested improvements below.

Suggested improvements: This paper will benefit from responses to the comments below.

Line 1: This is an aside and not a criticism, but it would be good if the authors can create a better name for severe ozone depletion than “ozone hole.” The public is right to be confused by this phrase. “Severe ozone depletion” or “severely depleted ozone layer” define the phenomenon much better than “hole,” for a literal “hole” through the entire ozone layer has never been observed from the surface or by satellite. (When total ozone fell to 216 DU a decade ago at my site, it never occurred to me to define that event as an “ozone hole”. It was simply a highly unusual dip in the ozone amount.)

Lines 9-10: The text reads: “Low-latitude volcanic super-eruptions can destroy the life-protecting ozone in [the] tropics and spawn a massive ozone hole.” The text is repetitious and perhaps overly cataclysmal. A condensed version might read: “Low-latitude volcanic super-eruptions can significantly reduce the life-protecting ozone in the tropics by greatly increasing solar UV-B.”

Lines 50-52: The sentence has no citation. Are refs. 5, 17 & 18 applicable? Or is another citation appropriate?

Line 77-79: Good points.

Line 112-121: This finding, if novel, is the highlight of the paper.

Line 128-130: How does this text relate to the apparently incorrect 5 DU ozone loss from Pinatubo in Fig. 5?

Line 140: The text defines AOD: “AOD, being the total sunlight extinction” Please revise to explain that AOD is the aerosol fraction of total optical depth. (OD is the total optical depth with Rayleigh scattering included.)

Line 148: The text reads: “. . . the aerosol optical depth at middle latitudes reached a maximum of >5 (Fig. 2, Fig. 3 and Methods). This means that the sky was pitch dark . . .” Assuming that the AOD is for the peak of human vision (~550 nm), the sky is not “pitch dark” when the AOD is 5. For example, on 30 Aug 1997 at Alta Floresta, Brazil, the AOD at 540 nm reached 5.43 at 15:21:08. The sky was not pitch dark and was more like dusk. The sun could be safely seen without sunglasses through the thick biomass smoke. (Mims, unpublished report to GSFC).

Lines 157-159: The text reads: “ during the peak AOD stage of the volcanic plume evolution, SO₂ was converted into the less efficiently UV-absorbing sulfate aerosols, leading to a partial offset of the ozone depletion and the associated UVI increase.” The “peak AOD stage” is given as “>5” (line 148 above). During the Brazil campaign described above, when the AOD at 550 nm reached 5.43, the UV Index measured by a calibrated Solar Light PMA2100 UV radiometer fell to 0.00. (Mims, unpublished report to GSFC). Please explain how modeled measurements transitioned to an “associated UVI increase.” Post Pinatubo data show a decline in total ozone and an increase in UV-B (mainly during 1992), but the increased UV-B was not catastrophic.

Line 170-206: The discussion leaves this reader confused about the contradiction of a volcanic winter accompanied by an ultra-high UV Index. Both phenomena are described as catastrophic in nature. Based on the prior text, the high UV levels follow the darkened (but not pitch dark) skies following the eruption. This needs to be succinctly expressed in the discussion.

Figure Captions: Might be best to add to the caption for Fig. 1 words to the effect that the figure is from GISS Model E (2006), which was also used for subsequent figures.

References: I assume that a paper about the presumed ozone impact on the historic and much more recent Samalas eruption was not addressed and cited because of its very recent publication date (Oct 2020). (The present paper does not mention the Samalas eruption—but the authors do cite the 2019 tropical eruption paper by Brenna et al. which does.) The authors should address this new paper and compare its ozone findings with theirs. The paper is: David C. Wade, et al., Reconciling the climate and ozone response to the 1257 CE Mount Samalas eruption. *Proceedings of the National Academy of Sciences* Oct 2020, 117 (43) 26651-26659; DOI: 10.1073/pnas.1919807117.

Clarity and context: The abstract is fine. The text and conclusions within the discussion are addressed above.

Please indicate any particular part of the manuscript, data, or analyses that you feel is outside the scope of your expertise, or that you were unable to assess fully: While I scanned the 2006 paper that describes the GISS Model E in detail, I made no attempt to implement the model.

Response to Reviews

Dear reviewers,

We thank you for your insightful comments, critical considerations, and help in improving the manuscript. In the following, we explain how we adjusted the manuscript to address your questions point by point.

Reviewer #1 (Remarks to the Author):

This is an interesting paper challenging the conventional idea of massive volcanic eruptions impacting life on Earth through the so-called volcanic winter. The authors point out the shortcomings of such a scenario. Using simulations with a state-of-the-art global chemistry-aerosol-climate model, the authors show that the Toba eruption should have led to severe stratospheric ozone losses in tropics and hence extensive increases in solar UV levels at the surface. This would have resulted in serious stresses for living cells with environmental and ecological consequences. Studies on the impact of future large eruptions on societies and ecosystems are currently focused on the volcanic winter scenario. The results show that they should also consider an ozone-UV scenario. This kind of papers challenging conventions and paradigms fits well in Nature. However, the text is a bit missing the novel point of the paper. For instance, the title is about Toba causing ozone losses. They claimed to have discovered a “previously unaccounted mechanism that influences ozone” (also pointed out in the abstract: “hitherto overlooked mechanism”). This ozone loss mechanism is not new, only the consequences for UV levels and the emerging of a volcanic ozone-UV scenario are new. I have to recommend publication but with major revisions. I am happy with the scientific content but there is still a lot of work to do on the text, i.e. correcting misleading claims and stressing the novel scenario.

Major points:

- The authors should tone down their claim of a discovery about a “previously unaccounted mechanism that influences ozone” (also pointed out in the abstract: “hitherto overlooked mechanism”). This mechanism has been accounted before.

They explain this mechanism in the text (L112-1125) and abstract: “But massive tropical volcanic eruptions such as that of Toba give rise to large amounts of SO₂ and sulfate aerosols in the stratosphere, which strongly enhance the UV extinction optical depth. The SO₂ cloud strongly absorbs UV radiation, and weakly absorbing sulfate additionally scatters the photons back to space, which decreases the radiation flux needed to form ozone within and below the volcanic plume.... Showing shutdown of the O₂ and strong reductions of the O₃ photolysis within the volcanic plume ”.

Bekki et al., Nature, 1993, decompose exactly this effect of SO₂ absorption of UV on O₂ photolysis, ozone production and resulting ozone losses for the Pinatubo case and Bekki, 1995

and Bekki et al., GRL, 1996, takes explicitly into account this mechanism of ozone losses for the Toba case.

Michelangeli et al., JGR, 1989, takes into account the UV scattering by volcanic stratospheric aerosols for the Pinatubo case.

We are aware of the excellent and pioneering work by Bekki, which for some reason was not referenced in the draft. We have corrected that and Bekki et al, 1993, 1996 and Bekki 1995 are now properly cited. Thank you for pointing this out!

We would like to explain our initial claim of the “unaccounted mechanism”. The work by Bekki and colleagues considered only the effects of SO₂ and did not include the effects of the sulfate aerosols, which appear to be equally important. We, however, were not aware of the work by Michelangeli et al., 1989, which indeed investigated the effects of the sulfate aerosols on the photochemistry including the photolysis of O₂. Following the reviewer’s advice, we adjusted the text accordingly.

- The authors could discuss a bit more the alternative ozone-UV scenario and possible gains on shortcomings of the volcanic winter scenario. In particular, they should stress more that the ozone-UV scenario is not limited to the effects of volcanic sulphur. The scenario also holds for volcanic halogens with several model calculations also showing massive ozone losses with volcanic halogens.

Thank you. We’ve edited the manuscript and highlighted the role of the volcanic halogens.

Minor points:

- L67: “While the “missing” sulfur source remains to be found to resolve this paradox”. The “excess” sulphur paradox is not a complete mystery. We know it is probably due to the presence of an exsolved vapor phase in the magma before eruption (see 23 and Wallace, 2003] but this process is poorly understood and quantified.

Thank you, we’ve added the Wallace, 2003, reference.

- L121-122: missing word: “...showing shutdown of the O₂”, add photolysis

Thank you, we corrected the text.

- L131-132 and also later: The fact that ozone anomaly was peaking and massive in the tropics was already shown in Bekki et al, GRL, 1996, in the same case (Toba).

Thank you, we’ve incorporated the missing citation into the manuscript.

- L187-191: Very important point that these simulations are lower limits regarding ozone losses. Volcanic halogens have been largely neglected in chemistry-climate simulations. Model

simulations converge on the effect of volcanic halogens on the ozone layer with massive ozone losses: Brenna, 2019, but also Klobas, 2017, and Cadoux, 2015. Therefore, the alternative ozone-UV scenario is not limited to volcanic sulphur but should also include the halogen component.

We agree that volcanic halogens further enhance the ozone-UV effects. Thank you for the references, they were added.

- The authors do not discuss stratospheric H₂O. First, it could be a limited factor in sulphur oxidation to sulphate. At the same time, the warming of the tropical tropopause (at least in models) leads to much more water entering the stratosphere and hence more ozone destruction by the very effective HOx cycles in the lower stratosphere where most of the ozone is found. What are the changes in stratospheric H₂O in their model simulations?

The HOx catalytic cycle is known to have an effect on ozone (acknowledged in our manuscript, lines 98-100) and the model indeed simulates the enhanced water transport across the weakened tropopause. Nevertheless, our analysis shows that the bulk of the ozone loss is due to radiative effects caused by SO₂ and SO₄ aerosols, which we are able to isolate by performing diagnostic sensitivity simulations with the radiative photochemical feedbacks of SO₂ and SO₄ aerosols switched off (feedbacks on the actinic flux calculations).

The overall effects of the stratospheric moistening (and tropospheric drying) are accounted for in our simulations. Please, refer to Figure A10 (top panel) in our previous publication <https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2019JD031726> for the changes in stratospheric water.

Reviewer #2 (Remarks to the Author):

Key results: The authors have quantified highly significant reductions in total ozone they believe accompanied the Toba eruption. Leaving aside their speculations regarding the effect of the eruption on the survivability of humanity, the major significance of this work is the increase in UV-B that might occur should a Toba-class eruption occur today.

Validity: The paper flaws that must be reasonably resolved. For example, the paper proposes both a sharp increase in UV-B resulting from major ozone destruction by Toba's emissions and their byproducts and significant blockage of UV-B by the volcanic plume. How the latter allows for the former is not clearly explained. Post-Pinatubo ozone, UV-B and AOD data are available. For example, see Fig. 5.5 at https://www.researchgate.net/publication/227860247_Surface_ultraviolet_radiation_Past_present_and_future/figures?lo=1 Is there not a role for actual measurements such as these in conjunction with model predictions? (This link is for 40N. Dozens of UVB Biometers and Dobson

spectrophotometers were in use at many locations when Pinatubo erupted.) At minimum, physical data could provide a test of model performance.

We agree that a vast amount of post-Pinatubo observations is available. Unfortunately, they are not applicable in this study. One of the key differences between the Toba and Pinatubo eruptions is the halogen loading in the atmosphere. The Pinatubo eruption in 1991 happened in the contemporary stratosphere characterized by the elevated levels of the anthropogenic chlorofluorocarbons and halons. The strong observed ozone depletion after Pinatubo eruption (to which reviewer also refers below) was due to heterogeneous chemical processing of the anthropogenic halogen species (for details and further reference, please refer to <https://agupubs.onlinelibrary.wiley.com/doi/epdf/10.1002/2017GL073972> or <https://www.nature.com/articles/srep12243>). The Toba eruption, on the other hand, happened in the pre-industrial atmosphere, free of anthropogenic halogens, and thus the impact on the ozone of the Pinatubo-size eruption (not Pinatubo, but eruption with equivalent SO₂ emissions) is smaller in our simulations. Please, note, that we explicitly state in the manuscript (lines 188-190), that simulations do not include the impact of halogen compounds, and thus our results present the lower boundary of the ozone depletion.

Given these considerations, unfortunately, we can not employ Pinatubo observations as additional validation to study Toba eruption, since the modeling setup for Pinatubo and Toba are drastically different.

Overall, with this explanation regarding the Pinatubo, we hope that the question about the validity of our modeling results (raised by the reviewer) is cleared.

Regarding the UV-B, please, refer to our reply in the “Validity of the approach” section below.

Originality and significance: The Toba eruption and its immense consequences have been reported in many papers. These papers do not always agree, and an umbrella statement to this fact might be appropriate, especially for those new to paleovolcanology. While some of these papers rely on studies and measurements of physical ejecta, others rely on model predictions and comparisons with more recent major eruptions (e.g., Tambora, Krakatau, Pinatubo, etc.). This paper falls in the latter category, with modeling forming its key analysis mechanism. I am unaware of prior work that gives claims for the magnitude of O₃ destruction and UV increase as specific as this paper. Assuming that the paper can be revised to accommodate concerns given below, their predictions should be considered by both atmospheric scientists and those who study the impact of UV-B on human health and the survivability of plant life.

Data & methodology: Figure 5 is both mistaken and troubling. (A) It is mistaken because it ascribes a loss of ozone following Pinatubo of only 5 DU. This is incorrect, for various papers have described much larger losses. For example, Grant, W. B., et al. (1994, *Aerosol-associated changes in tropical stratospheric ozone following the eruption of Mount Pinatubo*, *J. Geophys. Res.*, 99, 8197– 8211) reported O₃ losses of as much as 33%. (This reviewer measured 60-day average differences at 29.6N of pre-and post-Pinatubo spring O₃ of 26 DU (-14.8%) and winter

O3 of 32 DU (-17.8%). Paper submitted.) (B) Figure 5 is troubling, for no reference or citation is provided for the 5 DU assertion for Pinatubo. (Line 359 suggests that Fig. 5 is from the model. That should be stated in the caption unless the model is cited in the caption for Fig. 1 as suggested herein.) If the predicted O3 decreases for putative eruptions greater than Pinatubo are a product of the model(s) employed by the authors, Fig. 5 could raise questions about other results in the paper.

Please, refer to our reply in the “Validity” section above.

Please note that the purpose of Figure 5 is to show the scaling of the ozone depletion (under modeling assumptions representative for Toba eruption, free of anthropogenic halogens). The original caption reads “Pinatubo size eruption”, rather than actual “1991 Pinatubo eruption”. To avoid further confusion, we have improved the caption of Figure 5. Thank you for pointing this out.

Validity of the approach, quality of the data and quality of presentation: Concerns about Fig. 5 are given above. Another concern is the need for more discussion to better explain the increase in UV-B that will accompany their predicted O3 losses. The paper observes that UV-B that leaks through what remains of the ozone layer will be modulated both by the attenuation of the volcanic aerosol cloud and absorption by both O3 and SO2. But how the UV-B managed to achieve ultra-high levels in the presence of the post SO2 sulfate cloud was not fully explained.

We agree that the perturbed surface UV radiation can be confusing and difficult to understand. To remedy this, we have added more details into the manuscript regarding the UV index calculations (section “UV index”, after line 387).

Please note that elevated SO2 or SO4 aerosol concentrations do not simply cancel out the effects of O3 depletion. Their concentrations and effects on UV radiation change and evolve in time. The UV index is an excellent diagnostic to describe the overall effects of the perturbed O3, SO2 and SO4 (not just O3 alone) on surface UV radiation. In the revised manuscript we explain that during the initial stage the UV radiation is almost entirely blocked (with still unperturbed ozone column and maximum SO2 concentrations, line 157). During this stage the suppression of the O2 photolysis is strongest, but O3 depletion is not instantaneous and equilibration takes time (please refer to the Chapman mechanism for the ozone lifetime). As the volcanic plume evolves, the surface UVI near the equator increases due to depletion of the O3 (figure 1) and depletion of the SO2 column (figure 3, note the log scale) as it is oxidized into SO4. Approximately 9 months after the eruption, SO2 is almost entirely oxidized (10 DU compared to initial 350 DU) and O3 column depletion peaks around 40-45% (DU). The effective radius of sulfate aerosols also peaks in 9 months, reducing the efficiency to scatter the UV radiation (figure 2, <https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2019JD031726>). The enhanced SO4 loading only partially offsets the increased UVI due to ozone depletion. Note that visible sulfate AOD near the equator peaks at 3-3.5 (not >5). Together, perturbed O3, SO2, and SO4 combine to produce the largest overall increase in UVI in about 9 months near the equator.

During the later stage of the volcanic plume evolution, the surface UVI returns to the background level as SO₄ is removed from the atmosphere and O₃ recovers.

Appropriate use of statistics and treatment of uncertainties: The modeled charts in Figs. 1-4 do not include error bars, which is customary for visualizations. As for the model, the 2006 version of the GISS Model E is used in this paper. This model has been updated to GISS Model E2.2, a 2020 version that specifically advances results for elevations in the upper troposphere and stratosphere that are most impacted by Plinian volcano eruptions. The authors might want to look into this updated version of the model they used. Details are here: Rind, D., Orbe, C., Jonas, J., Nazarenko, L., Zhou, T., Kelley, M., et al. (2020). GISS Model E2.2: A climate model optimized for the middle atmosphere—Model structure, climatology, variability, and climate sensitivity. *Journal of Geophysical Research: Atmospheres*, 125, e2019JD032204. <https://doi.org/10.1029/2019JD032204>

One of the aims with the error bars is to show that the signal can be distinguished from the natural variability of the system. In our case (for example, Figure 1), ozone depletion or sulfate optical depth far exceed the natural variability. We think that error bars will not add information but rather make the figures less straightforward to read.

Thank you for the ModelE 2.2 reference. We aim to employ it in our future work.

Conclusions: Please see suggested improvements below.

Suggested improvements: This paper will benefit from responses to the comments below.

Line 1: This is an aside and not a criticism, but it would be good if the authors can create a better name for severe ozone depletion than “ozone hole.” The public is right to be confused by this phrase. “Severe ozone depletion” or “severely depleted ozone layer” define the phenomenon much better than “hole,” for a literal “hole” through the entire ozone layer has never been observed from the surface or by satellite. (When total ozone fell to 216 DU a decade ago at my site, it never occurred to me to define that event as an “ozone hole”. It was simply a highly unusual dip in the ozone amount.)

We agree with the reviewer and will adopt this reasoning in future work.

Lines 9-10: The text reads: “Low-latitude volcanic super-eruptions can destroy the life-protecting ozone in [the] tropics and spawn a massive ozone hole.” The text is repetitious and perhaps overly cataclysmal. A condensed version might read: “Low-latitude volcanic super-eruptions can significantly reduce the life-protecting ozone in the tropics by greatly increasing solar UV-B.”

Thank you. This is the requirement of the journal. Formatting will probably change and this sentence will be placed elsewhere during the typesetting.

Lines 50-52: The sentence has no citation. Are refs. 5, 17 & 18 applicable? Or is another citation appropriate?

Multiple citations 15,17,18,19 are provided and detailed in the next sentence.

Line 77-79: Good points.

Line 112-121: This finding, if novel, is the highlight of the paper.

Line 128-130: How does this text relate to the apparently incorrect 5 DU ozone loss from Pinatubo in Fig. 5?

Please, refer to our reply in the “Validity” section above.

Line 140: The text defines AOD: “AOD, being the total sunlight extinction” Please revise to explain that AOD is the aerosol fraction of total optical depth. (OD is the total optical depth with Rayleigh scattering included.)

Thank you for spotting this. We removed the misleading “total”.

Line 148: The text reads: “. . . the aerosol optical depth at middle latitudes reached a maximum of >5 (Fig. 2, Fig. 3 and Methods). This means that the sky was pitch dark . . .” Assuming that the AOD is for the peak of human vision (~550 nm), the sky is not “pitch dark” when the AOD is 5. For example, on 30 Aug 1997 at Alta Floresta, Brazil, the AOD at 540 nm reached 5.43 at 15:21:08. The sky was not pitch dark and was more like dusk. The sun could be safely seen without sunglasses through the thick biomass smoke. (Mims, unpublished report to GSFC).

We have also checked our dust storm calculations for the optical depth of 4.5 and found that at noon half of the SW flux reaches the surface. Text was corrected accordingly. Thank you for pointing this out!

Lines 157-159: The text reads: “ during the peak AOD stage of the volcanic plume evolution, SO₂ was converted into the less efficiently UV-absorbing sulfate aerosols, leading to a partial offset of the ozone depletion and the associated UVI increase.” The “peak AOD stage” is given as “>5” (line 148 above). During the Brazil campaign described above, when the AOD at 550 nm reached 5.43, the UV Index measured by a calibrated Solar Light PMA2100 UV radiometer fell to 0.00. (Mims, unpublished report to GSFC). Please explain how modeled measurements transitioned to an “associated UVI increase.” Post Pinatubo data show a decline in total ozone and an increase in UV-B (mainly during 1992), but the increased UV-B was not catastrophic.

As we explained in the manuscript, the maximum of the volcanic winter effects (cooling) is located in high latitudes, in the geographically different region from the maximum of the UV exposure in tropics. Similarly, the maximum of the sulfate AOD (>5) in the extra-tropics does not coincide geographically with the maximum of the high UVI in the tropics.

The abstract, main text and discussions explain that quite well. For example, we quote the abstract “An ozone hole in the **tropics** and volcanic winter in the **extra-tropics**” or line 148 “aerosol optical depth at **middle latitudes** reached a maximum of >5” and line 151 “near the **equator** ... 140% increase of ... UV index (UVI)”.

Figure 2 shows that the AOD stays below 3-3.5 (not >5) near the equator, i.e in the region of the max UVI increase.

Unfortunately, comparing the Brazil campaign (we assume that it is a biomass burning campaign) to post-Pinatubo observations is beyond the scope of our study (especially, without the details and exact numbers). We are also not aware of the technical details of the PMA2100 radiometer.

Below, we listed several considerations that could help in resolving the confusion.

1. Only a slight post-Pinatubo UV-B increase is consistent with our results, given that the ozone loss in 1992 was much smaller than after Toba (5% and 50% in tropics).
2. Conventionally, modelled UVI is reported for clear sky conditions during the local solar noon as the upper limit of the potential UV exposure. Given the strong UVI diurnal cycle, what was the solar zenith angle during the measurements? Was the cloud cover significant?
3. Unlike Toba or Pinatubo cases, the Brazil campaign is characterized by the unperturbed column O₃. Thus, zero UVI at the surface during the campaign seems consistent with the enhanced extinction in the atmospheric column (unperturbed column O₃ + additional brown carbon aerosols OD). Additionally, the brown carbon aerosols have much smaller SSA and absorb stronger, unlike the scattering sulfate aerosols, which increase the downward diffuse flux.

Please, refer to our reply in the “Validity of the approach” section regarding the overall UVI increase.

Line 170-206: The discussion leaves this reader confused about the contradiction of a volcanic winter accompanied by an ultra-high UV Index. Both phenomena are described as catastrophic in nature. Based on the prior text, the high UV levels follow the darkened (but not pitch dark) skies following the eruption. This needs to be succinctly expressed in the discussion.

Please, refer to our explanation above regarding the latitudinal distribution of the volcanic winter and UV effects. As we explain in the manuscript, these effects combine and are not mutually exclusive. We’ve edited the text and removed the misleading “pitch dark”. This should not cause confusion anymore. Thank you!

We would like to highlight that the aerosols optical depth is used in the text to define the location of the volcanic winter effects and strongest cooling, while we use the UVI to diagnose the

overall UV exposure. This latter diagnostic already includes the effects of the perturbed O₃, SO₂ and SO₄.

Figure Captions: Might be best to add to the caption for Fig. 1 words to the effect that the figure is from GISS Model E (2006), which was also used for subsequent figures.

Thank you, the caption was updated.

References: I assume that a paper about the presumed ozone impact on the historic and much more recent Samalas eruption was not addressed and cited because of its very recent publication date (Oct 2020). (The present paper does not mention the Samalas eruption—but the authors do cite the 2019 tropical eruption paper by Brenna et al. which does.) The authors should address this new paper and compare its ozone findings with theirs. The paper is: David C. Wade, et al., Reconciling the climate and ozone response to the 1257 CE Mount Samalas eruption. *Proceedings of the National Academy of Sciences* Oct 2020, 117 (43) 26651-26659; DOI: 10.1073/pnas.1919807117.

Thank you! The citation has been added in the line 107 and integrated into the discussion of the role of halogens.

Clarity and context: The abstract is fine. The text and conclusions within the discussion are addressed above.

Please indicate any particular part of the manuscript, data, or analyses that you feel is outside the scope of your expertise, or that you were unable to assess fully: While I scanned the 2006 paper that describes the GISS Model E in detail, I made no attempt to implement the model.

5th Feb 21

Dear Dr Osipov,

Your manuscript titled "Toba supervolcano eruption caused tropical ozone hole" has now been seen by our reviewers, whose comments appear below. In light of their advice I am delighted to say that we are happy, in principle, to publish a suitably revised version in *Communications Earth & Environment* under the open access CC BY license (Creative Commons Attribution v4.0 International License).

We therefore invite you to revise your paper one last time to address the remaining concerns of our reviewers. Please include a point-by-point response that takes into account the points raised. In particular, please ensure that you replace all reference to an ozone hole with "exceptional ozone depletion" or similar. At the same time we ask that you edit your manuscript to comply with our format requirements and to maximise the accessibility and therefore the impact of your work.

In addition, please consider the following points from us:

Line 60: "Analyses based on the recent melt inclusion technique suggested..."

The use of melt inclusions to reconstruct pre-eruptive volatile contents is no longer a recent development. I suggest rephrasing to "Pre-eruptive melt volatile contents, derived from melt inclusion analyses, suggest..."

Line 68: "While the "missing" sulfur source remains to be found to resolve this paradox..."

The discrepancy between petrological estimates of initial volatile content and remote sensing estimates of erupted volatiles, is widely recognised as indicative of the co-existence of melt with an exsolved volatile phase at depth (as indicated by Reviewer 1 during first review). Petrological estimates of initial sulfur content therefore underestimate the total erupted sulfur yield, particularly at hydrous arc volcanoes where an exsolved volatile phase is present from lower crustal depths. Please considering rephrasing the text in this section to reflect this.

e.g.

Wallace, P.J. and Gerlach, T.M., 1994. Magmatic vapor source for sulfur dioxide released during volcanic eruptions: evidence from Mount Pinatubo. *Science*, 265(5171), pp.497-499.

Wallace, P.J. and Edmonds, M., 2011. The sulfur budget in magmas: evidence from melt inclusions, submarine glasses, and volcanic gas emissions. *Reviews in Mineralogy and Geochemistry*, 73(1), pp.215-246.

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Please review our specific editorial comments and requests regarding your manuscript in the attached "Editorial Requests Table". Please outline your response to each request in the right hand column. Please upload the completed table with your manuscript files.

If you have any questions or concerns about any of our requests, please do not hesitate to contact me.

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We hope to hear from you within two weeks; please let us know if you need more time.

Best regards,

Emma Liu, PhD
Communications Earth & Environment
orcid.org/0000-0003-1749-9285

Joe Aslin
Associate Editor
Communications Earth & Environment

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors did a good job at responding to the comments but I still think that the paper needs to be improved before accepting it. Here are some comments using the "tracked changes" manuscript.

14: - "cause volcanic winters"

Rather: ...cause so-called "volcanic winters"

- 15: "We pose"

Pose is not right word. You propose

- "We show that stratospheric sulfate from the Toba supervolcano at 74 ka (thousand years ago) caused severe stratospheric ozone loss".

It is not only sulphate aerosols. It is also due to gaseous sulphur (SO₂).

Suggestion: we show that stratospheric sulphur emissions from...

73-74: "are the least conflicting among arguments that discredit the Toba catastrophe theory. Climate model simulations do not corroborate the initiation of glaciation".

No point exaggerating the volcanic winter scenario. Some people simply argue for a very strong and prolonged surface cooling without glaciation. Replace "the Toba catastrophe theory". By "the most extreme Toba catastrophe theories".

75-78: "The recent global climate simulations with improved representation of stratospheric chemistry and aerosol microphysics mechanisms have shown that the volcanic winter effects are significantly less extreme than assumed originally 11,12".

This point is more general. Add: "...the volcanic winter effects of very large volcanic eruptions are..."

References 11 and 12 are just model calculations. Stoffel et al., Nature, 2015 have demonstrated this point with model calculations AND observed climate proxies for the 2 biggest eruptions of the last millennium, Samalas and Tambora.

Stoffel, M., et al., Estimates of volcanic-induced cooling in the Northern Hemisphere over the past 1,500 years, Nature Geoscience, Nature publishing group, 8 (10), pp.784-788, 2015.

125: "...volcanic plume (Methods)".

Add reference 36 which describes it in detail.

200: "Our proposed stratospheric ozone and UV calamity"

Calamity is too colloquial.

203: "Our radiation attenuation mechanism"

It is not your mechanism, it has been proposed and studied in detail by others.

205: “We note the role of volcanic halogens, which have a similar outcome and enhance the ozone loss 21, 42, 43 . “

Similar outcome is not clear without a bit more information .

Suggestion: We note that, by neglecting volcanic halogen emissions, our simulations do possibly underestimate the extent of the stratospheric ozone losses. Indeed, volcanic halogens have a similar outcome and would enhance the ozone loss generating by volcanic sulphur 21, 42, 43 . “

Reviewer #2 (Remarks to the Author):

I’ve spent considerable time with this manuscript and note that the authors have made several revisions and corrections based on the two reviews. It’s especially good that the authors have added the Brenna et al citation, for that Nature paper covers much of the same territory Global ozone depletion and increase of UV radiation caused by pre-industrial tropical volcanic eruptions | Scientific Reports (nature.com).

The paper is now better prepared for publication, where it will join a family of papers that predict disaster should another Toba-scale eruption take place.

Because of my experience measuring the ozone layer for 31 years and being a former science magazine editor, I recommend just one major revision before the paper is accepted:

“Ozone Hole”: In response to my concerns about how the phrase “ozone hole” misleads the public and is technically erroneous, the authors write: “We agree with the reviewer and will adopt this reasoning in future work.” This is a puzzling response, for the authors have agreed with my concern but postponed enacting it. This problem begins with the title of their paper: “Toba supervolcano eruption caused tropical ozone hole.”

NASA’s web page on the Antarctic “ozone hole” begins with this definition: “The ozone hole is not technically a “hole” where no ozone is present but is actually a region of exceptionally depleted ozone in the stratosphere over the Antarctic that happens at the beginning of Southern Hemisphere spring (August–October).” Nasa Ozone Watch: Ozone hole facts

This definition is by Paul Nelson, who has been deeply involved in monitoring ozone from satellites and aircraft for at least 30 years. His phrase “exceptionally depleted ozone” is far more accurate than “ozone hole,” which continues to cause confusion in both the public and scientific audiences. If the authors do not wish to use the correct terminology, perhaps Nature’s editors can do so. I suggest the revision begin by revising the paper’s title to “Toba supervolcano eruption caused exceptional ozone loss.”

The comments that follow are merely suggestions regarding the author’s future work.

1. Pinatubo Comparison: I agree with the authors regarding how halogen did not play a significant role in the Pinatubo eruption. But I believe that eruption provides important clues about the transition from an optically thick atmosphere that significantly reduced UV-B to a clearer atmosphere in which highly reduced ozone allowed for unusually high levels of UV-B. (I measured this transition in detail and gave invited talks on this topic at NASA and a Japanese UV conference.) A figure illustrating a forecast for this transition is not required at this point but would greatly help this paper. In view of the importance of this topic, perhaps the authors can consider this for a future publication.

2. Tropical UV-B: I’ve measured UV-B in many locations since 1988. UV-B along the Amazon near the Equator is very similar to UV-B at my Texas site (29.6 N), and both these sites have much lower UV-B

then at sea level on Hawaii Island (when vog is not present). Why? Water vapor haze plays a significant role in modulating UV-B.

3. Raikoke Plume (2019): This does not relate to the present paper, but authors might like to know that the June 2019 eruption plume of Raikoke, possible the biggest eruption of this century so far, included up to 100 DU of HCl below 6 km. EGU2020-10651_presentation.pdf (copernicus.org) I made dozens of measurements of the elevation profile of the Raikoke plume over Texas and found no significant changes in the ozone layer or surface UV-B that could be attributed to that plume (which reached 22 km a few times at my site and over the Mauna Loa Observatory).

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Forrest M. Mims III

Response to Reviews

Dear reviewers and editor,

We thank you for your time and help in improving the manuscript. In the following, we explain how we adjusted the manuscript to address your questions point by point.

Reviewer #1 (Remarks to the Author):

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No point exaggerating the volcanic winter scenario. Some people simply argue for a very strong and prolonged surface cooling without glaciation. Replace "the Toba catastrophe theory". By "the most extreme Toba catastrophe theories".

Thank you. We are not trying to exaggerate the "Toba catastrophe theory", rather we refer here to the rejection of the entire "Toba catastrophe theory" (any interpretation, mild or extreme) by the paleoanthropological community. We first discuss that the paleoanthropological community considers mild 3.5 C cooling as insufficient and continue by introducing additional UV stress.

75-78: "The recent global climate simulations with improved representation of stratospheric chemistry and aerosol microphysics mechanisms have shown that the volcanic winter effects are significantly less extreme than assumed originally 11,12".

This point is more general. Add: "...the volcanic winter effects of very large volcanic eruptions are..."

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Suggestion: We note that, by neglecting volcanic halogen emissions, our simulations do possibly underestimate the extent of the stratospheric ozone losses. Indeed, volcanic halogens have a similar outcome and would enhance the ozone loss generating by volcanic sulphur 21, 42, 43 . "

Thank you, we edited the text. The lower limit of the ozone loss and UV increase is discussed in the previous paragraph (line 192).

Reviewer #2 (Remarks to the Author):

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Thank you, we edited the text and removed the “ozone hole” terminology from the manuscript, except when referring to the Antarctic ozone hole, for which this terminology is generally accepted, e.g. in textbooks. To comply with the reviewer comments, we have used hyphens for the expression in this context: “ozone hole”. The usage of this expression is also consistent with the NASA web page and Brenna et. al, referenced by the reviewer.

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We appreciate the discussion of the possible future work. We agree with the reviewer that Pinatubo eruption is a “gold standard” for the modeling studies and provides a wealth of observational data. Modeling various effects of the Pinatubo eruption is our concurrent research. We plan to carefully quantify the contribution of various mechanisms to the ozone depletion and will employ UV-B measurements. Thank you.

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Indeed, previous modeling efforts at most included clouds in addition to the volcanic plume itself. The drying of the troposphere and moistening of the stratosphere are well known post-eruption effects. The depleted ozone column can also amplify the effects of the water vapor on the UV radiation profile. Thank you for this interesting point.

3. Raikoke Plume (2019): This does not relate to the present paper, but authors might like to know that the June 2019 eruption plume of Raikoke, possibly the biggest eruption of this century so far, included up to 100 DU of HCl below 6 km. EGU2020-10651_presentation.pdf (copernicus.org) I made dozens of measurements of the elevation profile of the Raikoke plume over Texas and found no significant changes in the ozone layer or surface UV-B that could be attributed to that plume (which reached 22 km a few times at my site and over the Mauna Loa Observatory).

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Thank you again.

Forrest M. Mims III

Editor (Remarks to the Author)

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Thank you, we edited the text and added citations.