nature portfolio

Peer Review File

Global perturbation of stratospheric water and aerosol burden by Hunga eruption

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

6th Sep 22

Dear Dr Khaykin,

Your manuscript titled "Global perturbation of stratospheric water and aerosol burden by Hunga eruption" has now been seen by 3 reviewers, and I include their comments at the end of this message. They find your work on the impact of the Hunga eruption on the stratospheric water vapour of interest, but some important points are raised regarding the discussion of previous works and the use of the MLS version 5 data. 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.

We therefore invite you to revise and resubmit your manuscript, along with a point-by-point response that takes into account the points raised. In particular, please discuss previous works more thoroughly and state clearly the contribution of this study; clarify and discuss the use of MLS v5; and improve the structure of the result section. 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 weeks; 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 resubmission, 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,

Kerstin Schepanski, PhD
Editorial Board Member
Communications Earth & Environment
orcid.org/0000-0002-1027-6786

Clare Davis
Senior Editor
Communications Earth & Environment

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

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

This manuscript is generally well written and makes significant contributions to understanding of stratospheric water vapor (H2O) and aerosol perturbations caused by the 2022 Tonga volcanic eruption. It makes a thorough and compelling case that the injected stratospheric water vapor, both in its mass and its height of injection, is unprecedented in the satellite record and that it will likely cause long-lasting perturbations to atmospheric radiation and stratospheric chemistry. The synthesis of data from multiple instruments is novel compared to earlier works, including Xu et al. Atmosphere 2022 (Xu) and Millan et al. GRL 2022 (Millan), both of which focused primarily on MLS water vapor. This said, analysis of MLS water vapor in these earlier works should be cited more extensively throughout the manuscript when methods and results are similar to those previously published. Millan is cited in the body of the manuscript, but somehow fails to appear in the reference list. The only science results mentioned from Xu or Millan are their estimates of total mass of water vapor injected into the stratosphere. Both also discussed the zonal and meridional evolution of the plume, its rapid circumnavigation of the globe and its impact on radiative balance, albeit in less detail than the current work. Millan's figure S2 shows the consistency of the plume's vertical shear with reanalysis winds and ought to be mentioned in the discussion of Figure 3. Previous works also discusses the expected many-year impact of radiative forcing by the injected stratospheric water vapor from the Tonga eruption on climate as well as possible impacts on stratospheric ozone chemistry, and should be cited when these topics are discussed.

This work uses unscreened MLS version 5 water vapor, citing the previous use of unscreened MLS data by Millan. However Millan specifically recommended use of MLS version 4 water vapor. The retrieval phase that produces water vapor in v5, unlike that of v4, retrieves its own height-to-pressure profile; In the early days after the eruption when the retrieval is struggling with extraordinary profiles with elevated, spectrally interfering sulfur dioxide, this extra degree of freedom produces low height anomalies as large as 2.5 km relative to heights from other phases, and corresponding water vapor profiles with low anomalies that can be tens of ppmv relative to v4 values. These early anomalies likely do not significantly impact the results of this work, but (for example) MLS points on Figure 1c are likely 2-3 km low in height and the largest mixing ratios are likely ~30% low. These anomalies may also significantly impact the initial plume injection heights used in modeling studies. If the analyses are not repeated with v4 data, this should be discussed. At L127, where 'MLS' is first introduced, the data version should be specified.

The estimate of 9 days for the 30-40 km plume's circumnavigation time (L214) is faster than that estimated in Millan S2 (20-25 degrees of longitude per day is >13 days) or the 12 days reported by Xu, both of which used v4 MLS water vapor. The current work may focus on the leading edge of the plume, which could be reflecting blurring of higher-altitude, faster-moving plume signals by MLS averaging kernels. Altitude registration problems of v5 H2O may also play a roll. Again, the earlier results should be mentioned.

The analysis of this paper seems to be appropriate, statistically sound, and reproducible. I recommend that it be published after revision.

Reviewer #2 (Remarks to the Author):

Review to the article:
Global perturbation of stratospheric water and aerosol burden by Hunga eruption, by Sergey Khaykin et al.

Summary:
This paper describes the impact of the eruption of Hunga Tonga on the stratospheric composition with a view from different observations with a focus on aerosol and moisture. The authors display the formation and dispersion of the water and sulfate aerosol plume at a range from days to months by using satellite and ground-based observations and a simulation from the chemistry Lagrangian transport model CLaMS. This study complements and confirms the result of other publications on the Hunga Tonga eruption.

General comments:
The paper is written in fluent language and gives a detailed view on the Hunga Tonga eruption. I read the paper with great interest.
Before publication, the authors should address a few issues described below, and I recommend to restructure the paper to a certain extent.

1. The authors repeated results already published in other journals. Therefore, it is necessary that the authors state more clearly the new aspects in their analyses.

2. The text in the results section reads as a sequence of analyses of different data sets products and misses a clear story line. For instance, on page 4, the section: “Volcanic injection into the middle atmosphere” starts with the description of cloud top height, time evolution of cloud top height, then move on to the formation and decay of (parts) of the ice cloud, water vapour anomalies, sedimentation of ice, total water injection, motion and lifetime of volcanic ice.
Instead dividing the text into different stages of the volcanic cloud development, I would suggest to split the text into a part describing the development (vertical, zonal, meridional, of aerosol and moisture) and a part describing the internal volcanic cloud properties and processes (ice cloud, aerosol size and chemistry, water vapour, sedimentation of aerosol and ice).

3. The text is hard to read, because too often it is referred to figures in the supplement. I would recommend that the authors decide, which figures are necessary to describe their main and new aspects of the paper and present them in the main text and only refer to the supplement for additional information, that are not necessary for the reader to understand the main text.

4. A large amount of different data sets were used to describe the impact of the Hunga eruption on the stratosphere and described in the Method section. I would suggest to include a table listing the data sets and products for a better overview.

Page 8, Figure 2 (D,E,F): The colour chart is missing. The information gain from these figures is not clear.

Page 10, Figure 3: CLaMS figures should be larger displayed, otherwise the small circles are not visible.

Page 12, Figure 4: Why are there no iso-lines (of the CLaMS simulation) within the vertical cloud (blue)? Please explain, why CLaMS does no simulate water vapour anomalies in the northern lower branch of the BDC?

Reviewer #3 (Remarks to the Author):

Overall, this is a very complete and informative study of a unique event; certainly one that is of interest to the stratospheric aerosol, stratospheric chemistry, and global climate communities. The organization of the manuscript is clear and logical and tells a story about the Hunga eruption. The breadth of observations considered gives a complete picture of this event, which as you state “initiated a new era in stratospheric gaseous chemistry and particle microphysics”. First, I’ll outline a note about framing—the goal of your manuscript is to show that Hunga caused this new era and for this reason, there are a few places it would help to add comparisons to previous events to show why Hunga is unique. Next, I have a few notes about specific claims made in the introduction and the “volcanic injection into the middle atmosphere” section which are unclear. Finally, I’ll note that the final section of the paper, “global perturbation of stratospheric water and aerosol burden”, is the least convincing part of the paper. I point to a couple topics which are key to the understanding of volcanic aerosol plumes which aren’t mentioned here.

Because of your stated goal in the introduction, it would make sense to include more comparisons to previous stratospheric events as you go through the various observations. An example where you have done this well is lines 216-219, where you compare the magnitude of the eruption to the two largest volcanic events in the satellite record, Pinatubo and El Chichon. Comparison to other volcanic eruptions would help indicate how extreme and unique this event was and would help orient readers to the volcanic stratospheric injection literature. Specifically, I noted that lines 294-296 could be compared to Pinatubo literature concerning the “two branches” of cross-equatorial transport (see Aquila et al., 2012 “Dispersion of the volcanic sulfate cloud from a Mount Pinatubo-like eruption”, figure 4.). I have also noted that in the “Global perturbation of stratospheric water and aerosol burden” section, while you show that the Hunga eruption is certainly unique in the recent MLS record, it would be useful to understand if any of the perturbations seen in Figure 6a are a result of other stratospheric injections. You mention the Australian 2019/2020 wildfires in the text, for example.

The introduction and first section of the paper provide a great overview of the eruption and puts it in the context of the stratospheric injection literature. I have noted that your statement on lines 72-75, “Due to condensation near the cold point tropopause, moderately explosive eruptions of the last two decades only generated limited water vapor injections, in contrast with their substantial impacts on the stratospheric sulfur and aerosol budget”, seems to indicate that these conditions did not exist for the Hunga eruption. The cold point tropopause is still present during the Hunga eruption, as is shown in your figures, but the sheer mass of water injected during this eruption is what sets it apart from other recent eruptions. Next, your statement (lines 100-102) about eruptive column temperatures compared to ambient temperatures cites an example of a 1988 paper; is there a more recent modeling study you could reference here? On lines 123-126, you briefly describe a method for obtaining the total stratospheric water injection, but I am uncertain what you mean by “extrapolating the two early GNSS-RO profiles to the whole area of the young umbrella cloud and neglecting the remaining ice”. Would it be possible to add a more full description of this process? What do you mean by “neglecting the remaining ice”? Finally, on line 179, you mention Zhu et al. when mentioning that water may have increased the rate of sulfur dioxide oxidation in the Hunga plume; I think it is also worth mentioning that that paper, as well as Zhu’s other recent work, has also mentioned that heterogeneous oxidation of sulfur dioxide on the surface of ash as another consideration.

In the final section of the paper, I am left with a few remaining questions. First, on line 313, you state, “the extreme explosiveness of the Hunga eruption has led to in-depth perturbations of stratospheric gaseous and particulate composition”. While this is true, this would be true of every stratospherically relevant volcanic eruption or wildfire event. Your stated goal is to show that Hunga was unique and that it “initiated a new era”; it would make more sense to frame this in the context of the unique eruptive materials of Hunga Tonga, not just it's explosiveness. Next, you mention the gradual rise of the plume as a result of the BDC on line 323. There is also a well-documented effect of "self-lofting" in volcanic plumes where the radiative impact of volcanic gasses and aerosols in the plume can cause local heating, which results in a slow increase in altitude of the plume. Do you see evidence of this in your model or in the observations? Finally, you have a brief treatment of particle size in this section. While effective radius is a useful metric for particle size, it should be clear that this is not actually the size of the particles. Your analysis using effective radius is ample, just make sure that it's clear that you are using a summary statistic, effective radius, and not treating the actual size of the particles. My last note about particle size is that you only cursorily mention the ratio of water and sulfuric acid within the particles (lines 400-404). Do you see any evidence that because this is a uniquely humid situation, that some of the extra particle size is largely driven by extra water vapor on these particles?

Reply to Reviewer #1.

We thank the Reviewer #1 for the positive review and fair remarks, which have all been carefully implemented in the manuscript.

This said, analysis of MLS water vapor in these earlier works should be cited more extensively throughout the manuscript when methods and results are similar to those previously published. Millan is cited in the body of the manuscript, but somehow fails to appear in the reference list. The only
science results mentioned from Xu or Millan are their estimates of total mass of water vapor injected into the stratosphere. Both also discussed the zonal and meridional evolution of the plume, its rapid circumnavigation of the globe and its impact on radiative balance, albeit in less detail than the current work. Millan's figure S2 shows the consistency of the plume's vertical shear with reanalysis winds and ought to be mentioned in the discussion of Figure 3. Previous works also discusses the expected many-year impact of radiative forcing by the injected stratospheric water vapor from the Tonga eruption on climate as well as possible impacts on stratospheric ozone chemistry, and should be cited when these topics are discussed.
The discussion and citation of the previously published studies, in particular Millan et al. (2022) and Xu et al. (2022) that addressed the stratospheric water vapour perturbation and transport of the moist plumes have been enhanced. The manuscript now includes 10 citations of Millan et al. and other recently published studies (Xu et al., Schoeberl et al., Legras et al., Carn et al., Sellitto et al., Zhang et al; Voemel et al.) regarding the early evolution of the volcanic cloud, the effect of the vertical wind shear, the timescale of the meridional transport, the mass estimates as well as the unprecedented nature of stratospheric H2O perturbation and discussion of the possible impacts of the Hunga eruption on the stratospheric composition and climate.

This work uses unscreened MLS version 5 water vapor, citing the previous use of unscreened MLS data by Millan. However Millan specifically recommended use of MLS version 4 water vapor. The retrieval phase that produces water vapor in v5, unlike that of v4, retrieves its own height-to-pressure profile; In the early days after the eruption when the retrieval is struggling with extraordinary profiles with elevated, spectrally interfering sulfur dioxide, this extra degree of freedom produces low height anomalies as large as 2.5 km relative to heights from other phases, and corresponding water vapor profiles with low anomalies that can be tens of ppmv relative to v4 values. These early anomalies likely do not significantly impact the results of this work, but (for example) MLS points on Figure 1c are likely 2-3 km low in height and the largest mixing ratios are likely ~30% low. These anomalies may also significantly impact the initial plume injection heights used in modeling studies.
If the analyses are not repeated with v4 data, this should be discussed. At L127, where 'MLS' is first introduced, the data version should be specified.

As a matter of fact, we used both v4 and v5 in this study, however this was not properly explained in the manuscript. The analysis of the early plume evolution and particularly its vertical structure (which concerns Fig. 1c,d and Fig. 2 a,b,c) has been made with MLS v4 H2O/GPH following the recommendation by Millan et al. regarding the better accuracy of

height information in the v4 data. In Fig. 1c, the moist cloud tops at 52-53 km and the maximum water vapour mixing ratio is 145 ppmv, which is in full agreement with Fig. 1c in Millan et al. The usage of different versions of MLS data is now explicitly specified in the figures captions and discussed in the methods.

Regarding the impact of MLS data versions on the CLaMS CTM simulation: As mentioned in the supplement, we performed several simulations, initialized using both v4 and v5 data on different dates with and without QC screening. The results regarding the large-scale meridional dispersion of the plume (Fig.4, now Fig. 6) are largely insensitive to the data version used, however the simulations initialized by QC-filtered data showed significant discrepancies with the observations, which is why we have used the unfiltered data to initialize the simulation. The deficiency of meridional transport in CLaMS compared to MLS in the bulk plume layer is most likely due to large uncertainties in the imposed heating rates, which do not reflect the actual state that has been altered by the radiative effects of the excessive moisture. The respective sentence has been revised: "These differences between model and observations regarding meridional transport could be due to the sensor sensitivity limits in the upper stratosphere and/or due to uncertainties in initial injection height as represented in the model as well as due to uncertainty in the heating rates, which were altered due to radiative cooling in the stratosphere induced by excessive moisture (Sellitto et al., 2022)"

The estimate of 9 days for the 30-40 km plume's circumnavigation time (L214) is faster than that estimated in Millan S2 (20-25 degrees of longitude per day is >13 days) or the 12 days reported by Xu, both of which used v4 MLS water vapor. The current work may focus on the leading edge of the plume, which could be reflecting blurring of higher-altitude, faster-moving plume signals by MLS averaging kernels. Altitude registration problems of v5 H2O may also play a roll. Again, the earlier results should be mentioned.

The estimates of the circumnavigation timescales for different layers were indeed obtained by tracking the leading edge of the plume within the given layer. The timescale of 9 days corresponds to the uppermost part of the 30-40 km altitude layer and this timescale is confirmed by OMPS-LP aerosol tracking in Fig. 3b and in Taha et al., GRL, 2022. This timescale does not appear to depend on the data version – V4 and V5 analyses show nearly identical zonal progression rate using the same plume detection threshold. The text was revised as follows: "In the middle layer, between (30 - 40 km)(Fig. 4B), the aerosol and moisture plumes travelled in close tandem for about a month, and their leading edges circumnavigated the globe in 9 days, covering entirely the Southern tropical band by 29 January, that is in two weeks (Millan et al., 2022; Xu et al. 2022). Millan et al. (2022) showed the consistency of the plume's vertical shear with reanalysis winds."

All the technical remarks in the pdf file were implemented.

Reply to Reviewer #2.

We thank the Reviewer #2 for the appreciation and constructive remarks, which have all been carefully implemented in the manuscript.

1. *The authors repeated results already published in other journals. Therefore, it is necessary that the authors state more clearly the new aspects in their analyses.*

The following text was added to the introduction: " The January 2022 Hunga eruption on January 15, 2022 provided firsts observational evidence for significant volcano-driven stratospheric hydration (Millan et al., 2022; Xu et al., 2022), Without efficient sinks of moisture in the stratosphere, the ample hydration of this atmospheric layer is expected to persist for years, affecting various climatic variables such as stratospheric ozone(e.g., Dvortsov & Solomon, 2001), radiative balance (Millan et al., 2022; Sellitto et al., 2022; Zhang et al., 2022) and dynamics (Maycock et al., 2013). The global impact of the Hunga eruption on the stratospheric aerosol loading is another outstanding question required to assess the magnitude of climatic effects of this event.

With this study, we describe and quantify the stratospheric repercussions of the unique natural experiment in the middle atmosphere provided by the Hunga eruption. We investigate the formation and evolution of the stratospheric moisture and sulfate aerosol plume at a wide range of scales - from minutes and kilometers to monthly and planetary scales using a synergy of satellite and ground-based observations supported by transport modeling. Spanning nine months, the observations available to-date enable the first accurate assessment of the annual-scale stratospheric aftermath of this eruption, uncovering its climate-altering capacity."

The Discussion has been reworked to put forth the statement regarding the longevity of the perturbation based on the nine-month post-eruption evolution of stratospheric water and aerosol composition. In particular, the following text has been added: "The nine-month aftermath of the Hunga eruption in terms of stratospheric water vapour and aerosol burden, provided with this study, sheds light on the further evolution of this perturbation and its longevity. The stratospheric aerosol perturbation by the Hunga eruption has reached its peak in early June and exceeded in magnitude any volcanic or wildfire event in the last three decades. Since then, the stratospheric optical depth anomaly has been decreasing and assuming its further exponential decay, the extrapolation projects the return of SAOD anomaly to pre-eruption levels in 14-17 months, that is by mid-2023. We note that the gravitational settling of sulphate aerosols from the stratosphere may be expedited by their fast growth in the humid environment. The perturbation of stratospheric water vapour burden by 13% is tremendous and has no frame of comparison in the entire observation record dating back to 1985. As there are no efficient sinks of water vapour in the stratosphere, this perturbation is expected to last over several years. Indeed, in nine months since the eruption, the water vapour mass anomaly has only decreased by 2.5%, which should lead to the perturbation timescale of 3-4 years."

We have also added a new section "Perturbation of stratospheric water isotopic composition", which points out a strong and unique enhancement of heavy water isotopologues in the stratosphere, suggesting the sea water (and not the tropospheric moisture) as the primary source of the stratospheric hydration by Hunga.

2. The text in the results section reads as a sequence of analyses of different data sets products and misses a clear story line. For instance, on page 4, the section: "Volcanic injection into the middle atmosphere" starts with the description of cloud top height, time evolution of cloud top height, then move on to the formation and decay of (parts) of the ice cloud, water vapour anomalies, sedimentation of ice, total water injection, motion and lifetime of volcanic ice. Instead dividing the text into different stages of the volcanic cloud development, I would suggest to split the text into a part describing the development (vertical, zonal, meridional, of aerosol and moisture) and a part describing the internal volcanic cloud properties and processes (ice cloud, aerosol size and chemistry, water vapour, sedimentation of aerosol and ice).

The storyline of the paper was conceived to describe the event in chronological order and in "zoom-out" perspective ("at a wide range of scales - from minutes and kilometers to monthly and planetary scales…"). Eventually, some of the chapters have been extended with additional results, which may have disrupted the apparent flow of the story. This concerns, in particular, the sections "Volcanic injection into the middle atmosphere" and "Global perturbation of stratospheric water and aerosol burden". In order to improve the readability of the storyline, while preserving its general concept, we have revised the structure as follows:

The first section was renamed to "Eruptive column and water phase transition". The text has been reworked and condensed to avoid overloading the section".

The section "Global perturbation of stratospheric water and aerosol burden has been split into three sections: "Three-dimensional structure and evolution of stratospheric water and aerosol perturbation", "Evolution of sulfate particles" and "Global perturbation of stratospheric water vapour and aerosol burden"

3. The text is hard to read, because too often it is referred to figures in the supplement. I would recommend that the authors decide, which figures are necessary to describe their main and new aspects of the paper and present them in the main text and only refer to the supplement for additional information, that are not necessary for the reader to understand the main text.
We agree about the disproportional referencing to the supplement. We have reworked the text to reduce the referencing to the Supplement and moved some figures from the supplement (Fig. S1a, Fig. S2b, Fig. S9) to the main body. Fig. 1a has been moved to the supplement. The text and the captions have been adjusted accordingly.

4. A large amount of different data sets were used to describe the impact of the Hunga eruption on the stratosphere and described in the Method section. I would suggest to include a table listing the data sets and products for a better overview.

We have included a table listing all the data sets into the Methods section.

Page 8, Figure 2 (D,E,F): The colour chart is missing. The information gain from these figures is not clear.

Figure 2d,e,f panels have been revised. Instead of Himawari-8 RGB-Ash images they now display the extent of the hydrated plume derived from MLS and COSMIC-2 detections of wet anomalies in the middle stratosphere on the respective days.

Page 10, Figure 3: CLaMS figures should be larger displayed, otherwise the small circles are not visible.

CLaMS maps are now shown in a separate figure (Fig. 5) for better readability.

Page 12, Figure 4: Why are there no iso-lines (of the CLaMS simulation) within the vertical cloud (blue)?

The reason why there are no CLaMS isolines in Fig. 4 (now Fig. 6) is that the simulation was initialized with MLS data on 18 January (D+3), when the plume has already spread in the meridional dimension to a certain extent. The inner isoline thus represents the initial extent of the wet anomaly in the simulation. The respective mention has been added into the figure caption.

Please explain, why CLaMS does no simulate water vapour anomalies in the northern lower branch of the BDC?

The text has been modified and completed: "…the lack of the lower-branch northbound transport in the CLaMS simulation may be linked with the uncertainty in the heating rates, which were altered due to radiative cooling in the stratosphere induced by excessive moisture (Sellitto et al., 2022) and/or due to errors in the ECMWF meridional winds in the tropics (e.g. Podglajen et al., 2014)."

Reply to Reviewer #3.

We thank the Reviewer #3 for the appreciation and fair remarks, which have all been addressed in the revised manuscript.

Because of your stated goal in the introduction, it would make sense to include more comparisons to previous stratospheric events as you go through the various observations. An example where you have done this well is lines 216-219, where you compare the magnitude of the eruption to the two largest volcanic events in the satellite record, Pinatubo and El Chichon. Comparison to other volcanic eruptions would help indicate how extreme and unique this event was and would help orient readers to the volcanic stratospheric injection literature.

To address this remark we have included a new figure (Fig. 3) showing comparison of the Hunga and El Chichon plumes observed by lidars at La Reunion and Mauna Loa observatories. The text in the section "Early evolution of volcanic cloud" has been completed with the respective discussion: " The primary aerosol plume at 27-30 km altitude, overpassing La Reunion island on D+6 - D+7, was marked by an remarkably high aerosol optical depth (AOD) of 0.6 and scattering ratio (532 nm) reaching 280 (Baron et al., 2022), which to our knowledge represents the most intense stratospheric aerosol plume ever observed by ground-based lidars. Figure 3 compares early lidar detections of the Hunga and El Chichon plumes at La Reunion and Mauna Loa observatories, both located downwind of the respective volcanic eruptions (Barnes et al., 1997). While the maximum scattering ratio and AOD of the Hunga plume is observed already on D+4, the El Chichon plume reaches its maximum scattering of 181 and AOD of 0.81 much later, on D+40. This corroborates the hypothesis on the expedited conversion of SO2 to sulfate aerosols due to abundance of water in the Hunga plume (Zhu et al., 2022)."

The following text has been included into the Discussion section: "The stratospheric aerosol perturbation by the Hunga eruption has reached its peak in early June and exceeded in magnitude the strongest volcanic or wildfire events in the last three decades, including 2009 Sarychev eruption (Jegou et al., 2013), 2015 Calbuco eruption (Zhu et al., 2018), 2019 Raikoke eruption (Kloss et al., 2021) and 2019/2020 Australian Black Summer wildfires (Khaykin et al., 2020)."

Specifically, I noted that lines 294-296 could be compared to Pinatubo literature concerning the "two branches" of cross-equatorial transport (see Aquila et al., 2012 "Dispersion of the volcanic sulfate cloud from a Mount Pinatubo-like eruption", figure 4.).

Thank you for pointing out this study. We have included the following text: "The observed meridional transport pattern and timescale is very similar to that reported after the Pinatubo eruption with the faster lower-stratospheric and slower mid-stratospheric branches (Aquila et al., 2012). In both Pinatubo and Hunga cases, the transport into the opposite hemisphere was slower at all levels, with the lower stratospheric branch showing

vertical separation in the northern subtropics, likely due to confinement by the Summer monsoon anticyclones."

I have also noted that in the "Global perturbation of stratospheric water and aerosol burden" section, while you show that the Hunga eruption is certainly unique in the recent MLS record, it would be useful to understand if any of the perturbations seen in Figure 6a are a result of other stratospheric injections. You mention the Australian 2019/2020 wildfires in the text, for example. **The following text has been added into the new section "Perturbation of stratospheric water isotopic composition" It is worth noting that the 2019/2020 Australian wildfires, which caused a significant hydration of the stratosphere (Khaykin et al. 2020), did not lead to a measurable large-scale increase of water isotopic ratio in the stratosphere (Fig. 8B), which is another indication for the sea water as the main source of stratospheric hydration by Hunga.**

The following text has been added into the new section "Global perturbation of stratospheric water vapour and aerosol burden: "The other volcanic or wildfire events did not lead to a measurable increase in the global stratospheric water budget, although local enhancements in water vapour were observed by MLS following the 2017 Canadian wildfires (Millan et al., 2022). "

The introduction and first section of the paper provide a great overview of the eruption and puts it in the context of the stratospheric injection literature. I have noted that your statement on lines 72-75, "Due to condensation near the cold point tropopause, moderately explosive eruptions of the last two decades only generated limited water vapor injections, in contrast with their substantial impacts on the stratospheric sulfur and aerosol budget", seems to indicate that these conditions did not exist for the Hunga eruption. The cold point tropopause is still present during the Hunga eruption, as is shown in your figures, but the sheer mass of water injected during this eruption is what sets it apart from other recent eruptions. Next, your statement (lines 100-102) about eruptive column temperatures compared to ambient temperatures cites an example of a 1988 paper; is there a more recent modeling study you could reference here?

As we do not have any reliable information on the thermal structure of the eruptive column, we decided to remove the respective considerations from the paper. The following parts of the text have been removed:

" However, this stratospheric moistening conjecture had never been proven from observations. Due to condensation near the cold point tropopause, moderately explosive eruptions of the last two decades only generated limited water vapour injections, in contrast with their substantial impacts on the stratospheric sulfur and aerosol budget (Sioris et al., 2015; 2016).

"Numerical simulations of large eruptive columns (e.g. Woods, 1988) show that their temperature exceeds that of ambient air by hundreds of K at the tropopause. Thus, the initial plume has effectively bypassed the cold trap of the tropical tropopause and lower stratosphere."

On lines 123-126, you briefly describe a method for obtaining the total stratospheric water injection, but I am uncertain what you mean by "extrapolating the two early GNSS-RO profiles to the whole area of the young umbrella cloud and neglecting the remaining ice". Would it be possible to add a more full description of this process? What do you mean by "neglecting the remaining ice"?

The RO-based estimates of the injected water mass were obtained as described below. First, water vapor columns from 20 km to 50 km ASL are computed for the two early water mixing ratio profiles (Fig. S1) through vertical integration. Before performing that step, unphysical values in the profiles are replaced as follows: mixing ratio values corresponding to relative humidities over ice larger than 100% are replaced by the saturation value (computed with ECMWF temperature), negative values and values lower than 5 ppmv (arising due to uncertainties in the procedure, e.g. in ECMWF T) by a background of 5 ppmv. The two estimates of the water vapor column are then multiplied by the area occupied by the plume at that time (150,000 km2 according to the geostationary satellite), which leads to two estimates of the injected mass (75 and 140 Tg, respectively). Obviously, this calculation only takes into account the water vapor, to which GPS-RO is sensitive, neglecting the effect of the later sublimation in the stratosphere of the ice still present at the measurement time (i.e. assuming most of it will fall out). This approach also relies on the assumption that the profiles are representative of the whole area covered by the plume (neglecting heterogeneities), which should be realistic if, as suggested by the profiles, the whole plume seen from the geostationary satellites is at ice saturation.

Finally, on line 179, you mention Zhu et al. when mentioning that water may have increased the rate of sulfur dioxide oxidation in the Hunga plume; I think it is also worth mentioning that that paper, as well as Zhu's other recent work, has also mentioned that heterogeneous oxidation of sulfur dioxide on the surface of ash as another consideration.

We added the following sentence: "Another potential factor of expedited SO2 conversion to aerosols could be heterogeneous oxidation of SO2 on the surface of ash (Zhu et al., 2018; Zhu et al. 2022)."

On line 313, you state, "the extreme explosiveness of the Hunga eruption has led to in-depth perturbations of stratospheric gaseous and particulate composition". While this is true, this would be true of every stratospherically relevant volcanic eruption or wildfire event. Your stated goal is to show that Hunga was unique and that it "initiated a new era"; it would make more sense to frame this in the context of the unique eruptive materials of Hunga Tonga, not just it's explosiveness.

The respective sentence has been modified: "The extreme explosiveness of the Hunga eruption along with the submarine nature of the source has led to in-depth perturbations of stratospheric water vapour and aerosol amounts."

The following sentence has been modified in the Discussion: "The extreme explosiveness of the Hunga eruption and the submarine location of the volcano add up to the unprecedented

character, magnitude and the propagation timescale of the global stratospheric perturbation (Millan et al., 2022; Zhu et al., 2022; Legras et al., 2022; Carn et al., 2022; Schoeberl et al., 2022.; Voemel et al., 2022).”

Next, you mention the gradual rise of the plume as a result of the BDC on line 323. There is also a well-documented effect of “self-lofting” in volcanic plumes where the radiative impact of volcanic gasses and aerosols in the plume can cause local heating, which results in a slow increase in altitude of the plume. Do you see evidence of this in your model or in the observations?

The self-lofting has been widely reported for the carbonaceous aerosols injected into the stratosphere by wildfire-generated PyroCb storms. Among the volcanic eruptions, only the 2019 Raikoke eruption has been reported to produce a self-lofting plume, in which the heating was the result of the presence of absorbing ash and dynamical confinement preserving the plume at high concentration. The Hunga aerosol plumes did not present any heating anomalies but rather a cooling anomaly (Sellitto et al., 2022), furthermore they have been associated with cold anomalies revealed by radiosoundings (Voemel et al., 2022). The local radiative cooling by water vapour has led to deceleration of the tropical upwelling of the moisture plume (Legras et al., 2022; Schoeberl et al., 2022), whereas the aerosol plume has been shown to settle down with a rate consistent with the satellite-based estimates of their particle size.

Finally, you have a brief treatment of particle size in this section. While effective radius is a useful metric for particle size, it should be clear that this is not actually the size of the particles. Your analysis using effective radius is ample, just make sure that it’s clear that you are using a summary statistic, effective radius, and not treating the actual size of the particles.

We've added a brief explanation that the effective radius is calculated from the fitted lognormal and a reference to the more detailed analysis: “ The effective radius parameter is not measured directly, but calculated from fitted lognormal distributions to the SAGE III extinction spectra (Supplementary notes, SV). While the retrieved particle size is dependent on composition assumptions, there is insufficient information in the SAGE spectra to determine both size and if the relative fraction of water in the droplet makeup may be changing.”. Unfortunately, while measurements of changes to the aerosol composition would be highly beneficial, the effect of changes to particle size and composition are similar in their effect on the spectral signature of SAGE measurements. Therefore, we have chosen to retrieve the particle size, given a range of assumptions on the composition. A brief explanation of this has been added into the supplement.

My last note about particle size is that you only cursorily mention the ratio of water and sulfuric acid within the particles (lines 400-404). Do you see any evidence that because this is a uniquely humid situation, that some of the extra particle size is largely driven by extra water vapor on these particles?

The effective radius parameter is not measured directly, but calculated from fitted lognormal distributions to the SAGE III extinction spectra (Supplementary notes). While

the retrieved particle size is dependent on composition assumptions, there is insufficient information in the SAGE spectra to determine both size and if the relative fraction of water in the droplet makeup may be changing.

21st Nov 22

Dear Dr Khaykin,

Your manuscript titled "Global perturbation of stratospheric water and aerosol burden by Hunga eruption" 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. In particular, we ask that you provide robust support for claims regarding the transport of water vapour from the Hunga eruption into high latitude vortexes or, alternatively, tone down these claims to reflect the uncertainties in your analysis. Furthermore, please explain the basis on which the water vapour anomaly is estimated in more detail.

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.

Please note that it may still be possible for your paper to be published before the end of 2022, but in order to do this we will need you to address these points as quickly as possible so that we can move forward with your paper.

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REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have satisfactorally addressed the concerns raised in my first review.
My only further substantive reservation is with the discussion of the inconsistency between poleward transport of water vapor in the upper branch of the BDC seen in MLS observations and in CLaMS simulations. In lines 303-304 and Figure 6a, the authors note that MLS observations do not show the advance of 3 ppmv anomalies into the USLM high northern latitudes that are seen in CLaMS, and they suggest that this may be due to lack of sensitivity in MLS. MLS H_2O (v4 or v5) in the upper stratosphere has single profile precision of 5-6% (<0.4 ppmv) and should have no problem seeing 3 ppmv anomalies unless they are confined to extremely thin layers. Indeed, MLS H_2O is routinely used to show elevated upper stratospheric water vapor, such as that which descends into the winter polar vortices, and it has been used in studies of mesospheric water vapor, for example in showing correlation between saturation and PMCs. While transport in the LMS may have brought parcels from HTHH to high latitudes in both hemispheres, there is strong evidence from the sharp gradients at the vortex edge that HTHH water vapor did not enter the SH vortex (above the

subvortex) to directly influence polar processing in the 2022 SH winter. Given the MLS obserations in the NH upper stratosphere, it may be the case that high water vapor from HTHH does not get to NH high latitudes in time to be incorporated into the 2022-2023 NH winter vortex. The descrepancy between the model and the observations is of particular interest given the likely termination of MLS (and all other EOS) obserations in 2023. We certainly expect HTHH water vapor to influence polar processing in coming years, but we will be increasingly reliant upon models to interpret this impact.

Reviewer #2 (Remarks to the Author):

The authors have carefully looked through my comments and accordingly modified their manuscript. All my comments have been addressed.

There is only one point, where I still have a question:
In the discussion section you state:
"Indeed,in nine months since the eruption, the water vapour mass anomaly has only decreased by 2.5%,
which should lead to the perturbation timescale of 3-4 years." Can you please explain how you infer from the small decrease in water vapour anomaly in the first nine month to a pertubation time scale of 3-4 years?

Reviewer #3 (Remarks to the Author):

The authors have considered my comments and made corresponding changes to the manuscript. I appreciate the time that the authors put into responding to my comments in the rebuttal document and the additional citations made where necessary. I am happy to recommend

Reply to Reviewer #1.

The authors have satisfactorally addressed the concerns raised in my first review.

My only further substantive reservation is with the discussion of the inconsistency between poleward transport of water vapor in the upper branch of the BDC seen in MLS observations and in CLaMS simulations. In lines 303-304 and Figure 6a, the authors note that MLS observations do not show the advance of 3 ppmv anomalies into the USLM high northern latitudes that are seen in CLaMS, and they suggest that this may be due to lack of sensitivity in MLS. MLS H2O (v4 or v5) in the upper stratosphere has single profile precision of 5-6% (<0.4 ppmv) and should have no problem seeing 3 ppmv anomalies unless they are confined to extremely thin layers. Indeed, MLS H2O is routinely used to show elevated upper stratospheric water vapor, such as that which descends into the winter polar vortices, and it has been used in studies of mesospheric water vapor, for example in showing correlation between saturation and PMCs.

Indeed, given the MLS precision in USLM, a 3 ppmv anomaly should be readily detectable as long as the layer is sufficiently thick. However, as a matter of fact, the extratropical plumes were quite thin (less than 1.1 km) and quite weak, as can be inferred from aerosol lidar observations in Fig. S4. It is thus conceivable that given the MLS vertical resolution of more than 3 km in the USLM, the thin and weak layers of enhanced water vapour could be left undetected. The respective text has been modified as follows: "*The MLS observations show that within five months of the eruption, the hydrated plumes have spread in both directions from 65° S to 35° N but mostly within the bulk plume layer (20 - 30 km). The deep Brewer-Dobson transport is not captured by MLS, whereas the CLaMS simulation lacks the lower-branch northbound transport. The differences between the simulation and observations regarding the meridional transport could be due to various factors, such as the broad vertical resolution of MLS complicating the detection of thin layers; uncertainty in the model heating rates, which were altered due to radiative cooling in the stratosphere induced by excessive moisture (Sellitto et al., 2022) as well as due to errors in the ECMWF meridional winds in the tropics (e.g. Podglajen et al., 2014).*"

While transport in the LMS may have brought parcels from HTHH to high latitudes in both hemispheres, there is strong evidence from the sharp gradients at the vortex edge that HTHH water vapor did not enter the SH vortex (above the subvortex) to directly influence polar processing in the 2022 SH winter. Given the MLS oberations in the NH upper stratosphere, it may be the case that high water vapor from HTHH does not get to NH high latitudes in time to be incorporated into the 2022-2023 NH winter vortex. The descrepancy between the model and the observations is of particular interest given the likely termination of MLS (and all other EOS) oberations in 2023. We certainly expect HTHH water vapor to influence polar processing in coming years, but we will be increasingly reliant upon models to interpret this impact.

We do not have enough evidence for the entrainment of volcanic plumes by the Antarctic vortex, nor we can argue about any substantial amounts of moisture/sulfates in the northern high latitudes. Thus, we refrain from any speculations regarding the potential intrusion of volcanic material into the polar vortices. The following sentence has been removed: "The dispersion of sulfates to the Antarctic region has thus occurred before the polar vortex formation, and the Dumont d'Urville lidar measurements report stratospheric aerosol layers at the edge of the vortex in early June (not shown)."

Reply to Reviewer #2

The authors have carefully looked through my comments and accordingly modified their manuscript. All my comments have been addressed.
There is only one point, where I still have a question:
In the discussion section you state: "Indeed, in nine months since the eruption, the water vapour mass anomaly has only decreased by 2.5%, which should lead to the perturbation timescale of 3-4 years." Can you please explain how you infer from the small decrease in water vapour anomaly in the first nine month to a pertubation time scale of 3-4 years?
The projection of the decay period timescale was made by a simple linear extrapolation of the observed trend. The text has been modified to clarify this: "*Indeed, in nine months since the eruption, the water vapour mass anomaly has gradually decreased only by 2.5% (4.3±0.1% annual rate), which should lead to the perturbation timescale of over 3 years, assuming the further linear decay trend.*"

Reviewer #3 (Remarks to the Author):

The authors have considered my comments and made corresponding changes to the manuscript. I appreciate the time that the authors put into responding to my comments in the rebuttal document and the additional citations made where necessary. I am happy to recommend