Re-review of "Evidence of a Transient Ozone Depletion Event in the Early Hunga Plume Above the Indian Ocean" by Millet et al.

The manuscript has been substantially revised in response to referee comments. In general, the authors have done a good job in responding to the points raised by the reviewers, and the manuscript has been considerably improved. However, several new issues have been introduced through the revision process, some comments on the previous draft have still not been adequately addressed, and a few things that escaped my notice during the initial review have become more obvious now that the manuscript has been cleaned up. Although many of my comments on the revised draft are minor corrections that should be easy to deal with, I do have a number of more major substantive concerns that need to be addressed before the paper is accepted for publication.

General comment:

One major comment from my previous review that has not been resolved in the revised draft is the magnitude of the IASI ozone anomaly and its apparent discrepancy with MLS-based estimates. The authors have redone their analysis of MLS measurements and now find average negative anomalies in ozone of 0.7 ± 0.5 ppmv at 17–12 hPa and 0.6 ± 0.5 ppmv at 26–32 hPa. Unlike in the original manuscript, these anomalies are now barely significant at 1σ. However, the IASI analysis was also redone, and the maximum (and highly significant) TCO anomaly of 40.1 ± 4.8 DU is slightly larger than it was before. A stratospheric column ozone (SCO) anomaly is now also calculated; its maximum value (also highly significant) is 49.9 ± 4.7 DU. TCO / SCO anomalies of this magnitude will be met with skepticism by many readers. As demonstrated in my previous review, even an anomaly as large as 1 ppmv applied uniformly over the entire range from 40 to 1 hPa (the bulk of the stratospheric ozone layer) would not come close to producing an SCO anomaly of 50 DU. In their response, the authors state that "a direct comparison between IASI total column ozone measurements and MLS stratospheric ozone measurements is not appropriate, as the two instruments sample different atmospheric layers and use distinct observation geometries and techniques". This statement misses the point – ozone is ozone, no matter who is measuring it. For convenience, the plot included in my previous review was based on MLS measurements, but it did not depend on them – the same analysis could be done with any ozone profile. I encourage the authors to do such an exercise themselves – take an ozone profile (from anywhere), compute the SCO from it, and then calculate the SCO anomaly based on perturbations to that ozone profile of different amplitudes. This should give a sense of the magnitude and vertical extent of the perturbation necessary to bring about an SCO anomaly of 50 DU. I feel that some discussion about the credibility of the large column ozone anomalies estimated from IASI data – and their inconsistency with the MLS-based estimates – should be added to the text.

Specific comments and questions:

Both major substantive issues and minor points of clarification, wording suggestions, and grammar / typo corrections are listed together in sequential order through the manuscript. Line numbers refer to the "clean" version of the revised manuscript, not the tracked-changes file.

- Abstract: The authors need to be mindful that many readers will look to the Abstract to get a basic sense of the paper (and they may not go beyond that). Therefore the Abstract needs to do a much better job of summarizing the study and clearly highlighting its novel aspects. For example, the fact that this is the first presentation of IASI data in the context of the Hunga eruption should be emphasized here. In addition, the finding that the reduction in ozone appears to have been confined to two distinct layers associated with two separate aerosol clouds is one of the few new aspects of this study and should be more clearly articulated.
- L5: delete "The" in front of "Ozone"
- L9: Given the poor vertical resolution of IASI data, it is not really appropriate to refer to IASI and MLS "profiles" together in the same sentence. I suggest replacing "profiles" with "measurements" in this line.
- L11-12: The TCO result is not actually covered in the main body of the paper, but I think it should be – see my more detailed comments on this point below.
- L18: There is no need to define the acronym "UVR" as it is not used again in the manuscript.
- L34-35: such as that --> such as those; add a comma after "Calbuco (2015)"
- L44: Why is the word "implied" used here? Stratospheric ozone losses and radiative changes have been documented following volcanic eruptions, as noted in this manuscript.
- L52: clouds (PSCs) volume --> cloud (PSC) volume
- L71: An early paper discussing the influence of the excess humidity from Hunga in accelerating conversion of SO_2 to sulfate aerosols by Zhu et al. [2022, Comm Earth & Environ, 10.1038/s43247-022-00580-w] should also be cited for this point.
- L72: The paper by Sicard et al. has now been published, so the citation needs to be updated both here in the text and in the reference list. Moreover, other papers should also be cited for the Hunga-induced stratospheric cooling, such as those by Sellitto et al. (2022), Coy et al. (2022), and Schoeberl et al. (2022, GRL, 10.1029/2022GL100248).
- L75-83: I do not think that the listing of stratospheric chemical reactions has added useful information to this paper. I understand that in their comments on the previous draft one of the other referees suggesting discussing in more detail the influence of chlorine and sulfur compounds on stratospheric ozone, including showing some chemical reactions. But in response to that comment the authors have simply listed the set of "key heterogeneous reactions" given in the review paper by Solomon et al. (1999), with absolutely no accompanying text to put these reactions into context or give a sense of which ones are generally more important following volcanic eruptions. Zhu et al. (2023) and Evan et al. (2023, in the supplementary material), both already cited in the manuscript, provide a detailed description of the post-Hunga heterogeneous chemical reactions inside and outside the plume. In addition, Wilmouth et al. (2023, PNAS, 10.1073/pnas.2301994120) and Santee et al. (2023, JGR-A, 10.1029/2023JD039169) discuss the stratospheric chemical processing in the months following the eruption. Thus I feel that the authors would be better off deleting the material in these lines and simply referring readers to the lengthy explanations in those previous papers. If the authors want to retain these equations in the paper, then more in-depth discussion of how they are relevant needs to be added to the text.
- L88-89: It is not appropriate to say that Evan et al. "documented" a doubling of ozone loss via O_3+Cl – they merely reported the results shown by the modeling study of Zhu et al. (2023). Moreover, while the rate of that particular reaction did double, the rates of other reactions

changed by even greater amounts, so it is not clear why that one has been singled out. It would be better to make a more general statement that the rates of key reactions increased substantially, leading to the 5% depletion of stratospheric ozone over the Indian Ocean observed by Evan et al.

- L97: To avoid repeating "eruption", it would be better to say "impacts of Hunga on ozone".
- L97: Although the unique aspects of this study are articulated more clearly in the revised draft than they were initially, I think that it would help to add here something along these lines: "… post-eruption. The goal is not to elucidate the chemical mechanisms giving rise to the observed low ozone, as they were investigated in detail by Evan et al. (2023) and Zhu et al. (2023). Rather, the objectives of the present manuscript can be summarized …".
- L100: traversed by --> obtained within
- L104-106: The sentence "Satellite observations of ozone profiles and columns were exclusively acquired within this region, complementing the ground-based data while offering global coverage and regular monitoring" is problematic. It could be interpreted as saying that the satellites did not make measurements outside of this region, which is not only inaccurate but also potentially confusing since their global nature is mentioned. I suggest instead saying "This study focuses exclusively on satellite measurements acquired in this region."
- L139: using MLS data at level 2 and version 4 (v4) --> using version 4 (v4) MLS level 2 data
- L140-142: The implications of the two different approaches to obtaining instrument pointing information for the MLS data are unclear, and actually this detail is not of much interest for the average reader. It would be better to delete the two sentences devoted to this topic and simply state that the extraordinary enhancement in H_2O from Hunga degraded the accuracy of some of the v5 MLS data products in the first few weeks following the eruption.
- L144-145: For clarity, it would be better to rewrite the first two sentences of this paragraph as: "Following the recommendations of Millán et al. (2022), the MLS profiles for January 2022 are sourced exclusively from level 2 v4 measurements (Livesey et al., 2020). The MLS profiles are categorized as Hunga-influenced or non-influenced using criteria detailed in the next paragraph."
- L148: in any --> on any; to mean --> to the mean
- L152-153: All v5 ozone and water vapor profiles within a 5-degree radius of each of the January 2022 Hunga-influenced profiles were collected, regardless of the satellite's ascending or descending node --> All v5 ozone and water vapor profiles (on both ascending and descending sides of the orbit) within a 5-degree radius of each of the January 2022 Hunga-influenced profiles were collected
- L160-161: Assuming that I have understood correctly, for clarity change "locations showing high water vapor and a negative ozone anomaly" to "locations showing both high water vapor and a negative ozone anomaly".
- L162: 23 January --> 23 January 2022
- L164: Both profile groups --> The two profile groups
- L169-170: Although the authors' response letter makes it clear that v4 and v5 O_3 and v5 H_2O data were screened but v4 H_2O data were not screened, the manuscript itself is confusing on this point. First it is stated that "all quality flags … were used on the raw profiles (with the exception of the v4 H_2O profiles)". Then it is stated that "Only the v5 and v4 O_3 profiles were screened". These two statements are contradictory.

- L189-190: The statement “the altitude of the tropopause, as estimated by the instrument” implies that the IASI dataset includes a retrieval of tropopause height. Similar statements are made on L271-272 and in the Fig. 1 caption. Is that really the case, or is tropopause height taken from meteorological analyses? Please clarify and amend these statements as needed.
- L194-199: The new paragraph on IASI retrievals requires clarification on several points:
  - The “significant ozone perturbations” were seen in the ozone retrievals from UV-visible instruments, not in ozone itself.
  - spectral ranges of ozone and SO_2 do not overlap in the IASI ozone retrieval --> spectral ranges used for ozone and SO_2 in the IASI retrieval algorithms do not overlap
  - I do not understand what is meant by “ozone vertical variability” – given IASI’s very coarse vertical resolution, it might be better to omit the word “vertical” here.
- L209: top of the atmosphere irradiance --> top-of-the-atmosphere irradiance
- L214 & 215: near-real time --> near-real-time
- L237-238: In my original review I noted that the MLS ozone dataset has been very well validated and used extensively in prior studies. In fact, these data have been central to literally hundreds of scientific studies looking at regions all around the globe, including multiple papers by different groups examining the effects of Hunga on stratospheric ozone. Thus, the skepticism about their validity inherent in the statement “Prior to drawing any conclusions based on the MLS ozone profiles, it is essential to verify their agreement with precise local lidar observations during unperturbed conditions” is completely unwarranted. This language should be moderated. If indeed comparisons between MLS and Maïdo DIAL O_3 profiles have not been done previously, as stated in the response letter, then that represents a new contribution whose unique value should be articulated here.
- L239-240: Two points: (1) What does “all recovered profiles” mean? Why “recovered”? This word is used again in L258. (2) The phrases “within a 5-degree region around the lidar site” and “setting the inter-comparison radius to a maximum of 5°” are redundant.
- L240-242: First, these two sentences are also highly redundant and should be merged. Second, “both orbit types” should be “both sides of the orbit”. I recommend rewriting these sentences as “We averaged together MLS v5 ozone profiles from both the ascending and the descending sides of the Aura orbit, which have acquisition times near Reunion around 10:15 and 21:45 UTC, respectively. On the other hand, …”.
- L253: The statement “$O_{3\,MLS}(z)$ represents the MLS ozone value from averaging kernel at an altitude z” makes no sense. $O_{3\,MLS}(z)$ represents the retrieved MLS ozone value. The MLS averaging kernels were (or should have been) applied to the lidar data for this comparison.
- L257: at different layers --> in different layers
- L271: The comma after “retrieval” should be a semicolon.
- L293: assumed to be of 0.02 --> assumed to be about 0.02
- L299: Results show --> Results in Fig. 2b show; both instruments --> the two instruments
- L305: Add a pointer to Fig. 2a after “respectively”.
- L315: also increasing the standard variation --> which also increases the standard deviation
- L317-318: Again, the language used here – “MLS appears to be a suitable substitute for lidar data in studying ozone levels” and “… supports the use of MLS data across the region” – gives the impression that the reliability of MLS O_3 data for this purpose was in doubt. This wording should be toned down. I suggest at least adding “as expected” to the first phrase and simply

deleting the second one. In fact, the sentence about the representativeness of Reunion data for the Indian Ocean region works better logically without that statement. Also: strong agreement --> good agreement

- L320-326: Why is this discussion of the ozone annual cycle of relevance for this paper? If this information is needed to help interpret any results shown here, then that needs to be made clear; otherwise, this text seems to be a pointless digression.
- L330: The authors state that they compared two datasets, but actually they made two sets of comparisons involving four different datasets altogether: MLS vs DIAL and IASI vs SAOZ.
- L333: The panel titles in Fig. 4 ("MLS & Lidar comparison" and "IASI & SAOZ comparison") give no hint of which way the subtraction goes, nor does the figure caption make it clear. In the text, the results are characterized as "MLS–DIAL" and "SAOZ–IASI". Please clarify whether these differences are "spaceborne" minus "ground-based" data or vice versa; also, if they are not already, make the two sets of differences consistent in terms of direction (i.e., to be parallel with MLS–DIAL, the TCO differences should be taken as IASI–SAOZ, not SAOZ–IASI).
- L340: with higher and --> with larger biases and
- L344: I find this discussion of the relative bias between MLS and DIAL O_3 confusing. It is stated that "the bias decreases to 0.24 ± 2.12%" from 40 to 45 km. However, to me it looks like the bias goes from roughly +0.5% at 40 km to nearly –1% at 45 km; that is, the relative bias grows in magnitude but changes sign over this altitude range. I do not see where the quoted value of 0.24% comes from.
- L345: difference and error --> relative bias and standard error
- L347: bias of ozone --> bias in the ozone
- L349-350: Note also that the increased difference and error at altitudes lower than 20 km may be due to the reduced satellite accuracy and precision --> Note also that the increased relative bias and standard error at altitudes below 20 km may be due to the reduced satellite accuracy and precision at those levels
- L353-354: Here again I am confused by the wording of the text. It is stated that "MLS profiles tend to slightly under-estimate ozone concentrations relative to DIAL". But the relative bias shown in Fig. 4a is positive through most of the vertical domain; since the differences were characterized as "MLS–DIAL" on L333, those results indicate that MLS over- (not under-) estimates DIAL concentrations. This needs to be clarified.
- L356-357: The relative dispersion RMSD=3.26% for the IASI/SAOZ comparison is characterized as "very low". But for the MLS/DIAL comparison, RMSD=1.27%. Why was that value described a "low" (L354) while the larger value for IASI/SAOZ is "very low"?
- L360: plume (25-30 km) being --> plumes (25-30 km) that are
- L364: "it appears relevant to use" is very odd wording. I suggest simply saying "we now use".
- L367 & Fig. 5: The figure has been greatly improved. However, it is virtually impossible to see the red contours on the SCO panels (a1-a9), where they could be mistaken for very high DU values. It would be better to simply omit the total SO_2 contours from those panels and amend the text in this line accordingly.
- L369: the selection criterion --> the Hunga-influenced selection criterion
- L370-371: I have two comments about "reveal an east-to-west displacement of both plumes … supports previous studies": (1) it's not clear what "both" means here. This word immediately follows "H_2O and ozone anomalies", so the reader naturally associates it with

those two quantities, but the deficit in ozone does not constitute a “plume”. I assume that SO_2 and H_2O are meant. In any case this needs to be clarified, perhaps by saying something along the lines of “Hunga-affected air masses” instead. (2) The east-to-west displacement of the Hunga plume is not a new result “revealed” by Fig. 5. It was reported in several previous studies, including Millán et al. (2022), Khaykin et al. (2022), and others. As the authors noted in their response, the prior studies did not specifically talk about ozone. Nevertheless, they did identify the movement of the Hunga plume, so they should be credited here; the vague allusion to “previous studies” is not sufficient.

- L372: Three points: (1) It would be better not to repeat “rapid” in this line; (2) influence of H_2O --> influence of excess H_2O; (3) Zhu et al. (2022) could also be cited for the rapid conversion of SO_2 to sulfate.
- L374: illustrating --> suggesting
- L375: the Hunga --> Hunga
- L381-382: The last sentence in this paragraph essentially repeats what was said in L372-373. The repetition is confusing since the reader is expecting new information to be conveyed.
- L384: record anomalies of –49.9 ± 4.7 DU were recorded 76.5°E --> a record anomaly of –49.9 ± 4.7 DU was measured at 76.5° E
- L385-386: this IASI SCO anomaly is more than 14 times below the average variability --> the magnitude of this IASI SCO anomaly is more than 14 times larger than the climatological variability
- L386: anomaly map … suggests --> anomaly maps … suggest
- L390: emphasize --> indicate
- L395: Two points: (1) selected by the criterion --> identified by the selection criterion; (2) it would be good here to remind readers what the two groups of Hunga-influenced profiles are.
- L400: by one of --> by each of
- L403: highest --> higher-altitude
- L405: lowest --> lower-altitude
- L406-407: ozone reduction in --> low ozone at
- L407-409: Presumably the ozone anomalies for the two clouds stated in absolute units (ppmv, DU/km) in L404 and L406 are computed from the MLS climatology. It is then a bit jarring to have another set of ozone anomaly values for the two clouds relative to the average lidar profile given in percent terms. This approach precludes easy comparison of the magnitude of the ozone anomalies based on MLS climatology with those based on lidar data. The MLS-climatology-based anomalies should also be quoted in terms of percent. Moreover, it is not clear why the anomalies calculated by differencing the Hunga-influenced MLS profiles and the average lidar profile are emphasized over those based purely on MLS data.
- L408: highest --> higher; this change should be reflected in the Fig. 6 legends as well
- L409: lowest --> lower; this change should be reflected in the Fig. 6 legends as well
- L409: coherent with Evan et al. (2023) who --> consistent with those of Evan et al. (2023), who
- L413: Two points: (1) “confirms previous research” – both Evan et al. (2023) and Zhu et al. (2023) should be explicitly cited here; (2) “the ozone anomaly is linked to a reduction of the

ozone layer": by definition, a negative anomaly is a reduction – what is at issue here is the cause. It would be better to say "the ozone anomaly arose from chemical loss".

- L415-422: Some acknowledgment that the results of this trajectory investigation are further confirmation of the passage of the Hunga plume over Reunion as established by Baron et al. (2023) and Evan et al. (2023) is needed in this paragraph; i.e., those papers should be cited.
- L420: trajectories simulation --> trajectory simulation
- L426-427: As with the Abstract, the authors should bear in mind that many readers will skip most of the detailed discussion in the text and jump straight to the Conclusions. Therefore the Conclusions section needs to do a better job of summarizing the study and identifying its novel aspects. For example, the last sentence of the first paragraph could be amended to better capture the diversity of measurements used: "… using IASI, MLS, and OMPS satellite observations, in conjunction with ground-based measurements from Reunion". The fact that this is the first presentation of IASI data in the context of Hunga should also be emphasized.
- L431: was passing --> passed
- L434: levels --> abundances
- L436: "TCO" and "SCO" should be redefined in the Conclusions or just written out. In addition, I find it strange that the TCO result is considered sufficiently important that it is highlighted in the Conclusions (and the Abstract, as noted above), yet was relegated to an Appendix. I come back to this point below.
- L437: indicated --> indicated that
- L437-440: The final sentences in the manuscript are not well composed. In my opinion they could be rewritten to better convey the message: "Hunga-influenced MLS profiles show a significant reduction in ozone over the 30–12 hPa pressure range. Ozone depletion occurred in two distinct layers, associated with two separate sulfate aerosol clouds. Within the higher-altitude (17.78–12.12 hPa) aerosol cloud, ozone decreased by an average of 0.7 ± 0.5 ppmv (1.1 ± 0.7 DU/km). Within the lower-altitude (31.62–26.10 hPa) aerosol cloud, ozone decreased by an average of 0.6 ± 0.5 ppmv (1.7 ± 1.4 DU/km)."
- L440: The paper ends rather abruptly. In addition to rewriting the last few sentences as suggested above, the authors should consider adding some sort of final sentence to put their results into context. For example, they could say something about how their finding that the observed ozone reduction appeared to be confined within two distinct aerosol layers adds new perspective to the studies that had previously reported chemical ozone loss in the week following the eruption.
- Appendix A: Although I do not disagree that Fig. A1, while helpful, is the sort of ancillary material that belongs in supplementary information rather than the main body of the paper, I am less convinced that that is true of the accompanying text. To me, if a result is sufficiently noteworthy to report in both the Abstract and the Conclusions, then it should be discussed in the paper itself, not just in an Appendix. I was struck by this when I got to the Conclusions and found a number (for the max TCO anomaly) that I had not seen in reading the paper. I feel that the TCO information in this short paragraph should be integrated into the discussion in Section 3.4 (which can still refer to Fig. A1, as it already does now).
- L444: anomalies, both in --> anomalies in both
- L445-448: Clarification is needed in several places in these lines:

    - this IASI TCO anomaly is more than 3 times below the climatological variability --> the magnitude of this IASI TCO anomaly is more than 3 times larger than the climatological variability
    - this anomaly is about 5 times below the variability --> the magnitude of this anomaly is about 5 times larger than the variability
    - When I first read this paragraph, I thought that it contradicted what was stated earlier. If this paragraph is kept in the Appendix (i.e., if the authors choose not to integrate it into the main text as suggested), then to reduce the possibility of confusion, I suggest adding this sentence at the end: "As discussed in Section 3.4 in the main text, the magnitude of the anomaly in SCO exceeds the climatological variability to an even greater degree."
- L459-472: The authors may wish to review the Acknowledgments carefully – there are some typos and missing words.
- L489-490: The second entry for Baron et al. (2023) appears to point to the preprint of a paper that has now been published and thus should be deleted.
- L650-652: As noted earlier, the paper by Sicard et al. has now been published, so the citation needs to be updated.
- L673: The Earth observing system microwave limb sounder (EOS MLS) on the aura Satellite --> The Earth Observing System Microwave Limb Sounder (EOS MLS) on the Aura satellite
- L680: 2018 --> 2022
- Figure 2 caption: between 2003 to 2021 --> between 2003 and 2021
- Figure 3 caption: average … profile from 2013—2021 observations at --> average … profile calculated from observations taken over 2013–2021 at
- Figure 4 caption: The solid red and dashed blue lines in panel (b) should be explained.
- Figure 5 & caption: Two points: (1) the selection criterion --> the Hunga-influenced selection criterion; (2) As noted above, the red SO_2 contours should be omitted from panels (a1)-(a9).
- Figure 6 caption: highest and lowest --> higher-altitude and lower-altitude
- Figure 7 caption: for the 23.5 km --> for the trajectory ending at 23.5 km