Reviewer's comment 2 (RC2)

This is an excellent and comprehensive paper that shows that stratospheric aerosol parameters can be retrieved from nadir backscatter measurements for a large volcanic plume that is above the ozone layer. The paper is well written, and the problem is approached systematically with detailed explanations and appropriate appendices. The paper is highly appropriate for the scope of AMT and needs very little revision for publication. A few minor comments are included below for the authors to address in a revision.

We sincerely thank the reviewer for the thoughtful review. We address each point raised in the detailed responses below.

RC2-Q1. The definition of wet aerosol mass (in the abstract) should be clarified (and why is "wet" in quotations?)

Answer: We agree and removed quotation marks in the revised manuscript. We have clarified that wet aerosol mass refers to the aerosol column mass of aqueous sulfate droplets, i.e., including water uptake at stratospheric temperatures and relative humidities. The altered text is as follows:

Abstract-Page 2, Lines 45–46: "We estimate the total Hunga stratospheric wet aerosol mass (sulfate solution droplets, including water uptake) to be $M_{aer} \sim 0.5 \pm 0.05$ *Tg."*

Page 24, Lines 659–660: "~ a corresponding wet aerosol mass (~0.47 Tg), where "wet" denotes aqueous sulfate droplets including water uptake at stratospheric relative humidities."

RC2-Q2. Section 3.1 Normalization to background radiances includes a correction for ozone profile differences but it is highly feasible that air density differences are not characterized by M2-SCREAM in the early plume. What is the potential impact?

Answer: We agree that M2-SCREAM may not fully represent plume-related density changes in the first days after the eruption. In principle, this could affect Rayleigh scattering (Bodhaine et al., 1999) and the scaling of ozone absorption (Guo and Lu, 2006), since both depend on air density. These effects could add some uncertainty to the normalized radiances. To this end, we have added the following paragraphs to the paper, and added the reference to (Guo and Lu, 2006).

However, during the course of this research, we performed a study to investigate the influence of variations in atmospheric density profile and ozone profile for different scenes characteristic of the period following the eruption. For a given set of TROPOMI BUV radiances, the following tests were done:

- *the original atmospheric temperature and pressure profiles of the accompanying scene were replaced while holding the ozone profile fixed;*
- *the original atmospheric ozone profile of the accompanying scene was replaced while holding the temperature and pressure profiles fixed;*
- *the original set of all three were replaced to observe their combined influence.*

For each of these combinations, several retrievals were done to observe the influence of these variations on the retrieval results. Replacement of the ozone profile was found to generate the greatest influence, and therefore we paid much more attention to defining this profile for each TROPOMI scene. However, differences in atmospheric density profiles due to changes in temperature and pressure were found to generate small effects (on the order of a few tenths in retrieved AOD and sub-kilometer changes in peak height).

The authors added the following sentences in the revised manuscript:

Page 12, lines 275–278: "We also examined the potential impact of possible air-density variations that may not be fully captured by the M2-SCREAM reanalysis during the early days after the eruption. Such differences could influence Rayleigh scattering (Bodhaine et al., 1999) and the scaling of ozone absorption (Guo and Lu, 2006), since both depend on air density."

Bodhaine, B. A., Wood, N. B., Dutton, E. G., & Slusser, J. R. (1999). On Rayleigh optical depth calculations. Journal of Atmospheric and Oceanic Technology, 16(11), 1854-1861.

Guo, X., and Lu, D.: Feasibility study for joint retrieval of air density and ozone in the stratosphere and mesosphere with the limb-scan technique. Applied optics, 45(35), 9021-9030, 2006.

RC2-Q3. Section 3.4 Description of the unimodal particle size distribution parameters does not make sense. What is the sensitivity to the choice of 8 discrete ordinates with delta M scaling?

Answer: Regarding the PSD used for the volcanic aerosol, we have corrected the description of the unimodal lognormal size distribution parameters (with the width/variance parameter now properly given as dimensionless).

Regarding the number of discrete ordinate streams used in the RT computations, we remark that it is not 8 discrete ordinates in total that is being used, but rather "8 ordinates in the polar half-space" (i.e. 16 streams in total between local nadir to zenith). We have changed the text to clarify this distinction. However, even with the 16 total streams and the delta-M scaling ansatz that were actually used in this work, a comparison was made against using 32 total streams with delta-M scaling (often considered a kind of "gold standard" in these types of retrieval settings) and these yielded negligible differences in spectra.

RC2-Q4. Section 3.5 No results or analyses are shown from the validation with synthetic data. Typically a retrieval result and comparison with the input state would be shown just to verify the algorithm fidelity.

Answer: To help validate the Hunga-Tonga (HT) retrieval algorithm forward model (FM), we did some initial testing by comparing simulations using our FM with calculations made by the OMI simulator model, for a case with typical atmospheric and surface conditions experienced following the HT eruption during the period of this study (that is, profiles of temperature, pressure and ozone, vertical characterization of the volcanic aerosol, plus aerosol PSD and surface albedo). The resulting spectra were found to be in good agreement. It should be noted that this test actually helped clarify some HT-FM input issues that were addressed at an earlier phase in the study.

We also performed some closed-loop tests on the HT retrieval algorithm itself, again for typical post-eruption scenarios encountered in this study. Here, two types of tests were done to test retrieval fidelity, each test based on synthetic spectra generated by the forward model using a pre-defined state vector $X_{true} = \{AOD, Pk\ Hgt\}$*.*

- *The first retrieval test used the HT-FM-generated synthetic spectra with no noise; this is to check that the algorithm does indeed retrieve the known state (i.e.* $X_{ret} \approx X_{true}$*). This test was successful.*
- *The second retrieval test again used the synthetic spectra, but this time including noise levels representative of TROPOMI UV Band 1 measurements; this is to check that the retrieval algorithm can again basically retrieve the known state (i.e.* $X_{ret} \approx X_{true}$*), but this time observing increases in estimated uncertainty and/or bias. This test was also informative and successful.*

The authors added more details of synthetic test in Section 3.5 as below:

Page 17, lines 466–474: "To further verify the fidelity of the retrieval algorithm itself, we performed a set of closed-loop validation tests using synthetic radiance spectra generated by the forward model. Each test used a pre-defined state vector $X_{true} = \{AOD, Z_p\}$ *to represent typical post-eruption conditions. Two cases were examined: (1) a retrieval based on noise-free synthetic spectra to ensure that the algorithm could reproduce the known state (*$X_{ret} \approx X_{true}$*), and (2) a retrieval using spectra with added random noise levels representative of TROPOMI UV Band 1 measurements to assess retrieval robustness under realistic conditions. In both cases, the retrieved parameters converged closely to the true inputs, demonstrating that the Hunga retrieval framework is stable and internally consistent. The noise-added tests also showed slightly larger retrieval uncertainties, as expected, but without systematic bias in AOD or* Z_p*. Overall, the results increased confidence in the forward and inverse model configurations used in the actual Hunga plume retrievals."*

In this regard, comments have been added to the end of both sections 3.2 and 3.3 to mention that these kinds of integrity tests were performed. However, we feel that, given the number of figures already present in the paper dealing with uncertainty and/or validation, it was not necessary to present an additional figure or figures specifically related to this question of retrieval integrity.

RC2-Q5. Section 4.2 The retrieved AOD varies greatly with the refractive index assumptions. Is this wide range of refractive indices realistic for Hunga?

Answer: We acknowledge that variations in the real part of the refractive index between the extreme values n=1.39 (~30wt%) and n=1.47 (~80wt%) have a large effect on the retrieved AOD values, i.e., Figure 8 (bottom).

As to whether this wide refractive-index range is realistic for Hunga, we have carried out some further analysis of this issue and added new material. In the revised paper, we have added a new Appendix F, in which GEOS-CCM–CARMA diagnostics (Case et al., 2023) show H_2SO_4 wt% ~40–70% on 17 Jan (core ~40%). This composition span is consistent with laboratory refractive-index measurements for sulfuric-acid solutions near 312 nm (e.g., Beyer et al., 1996), and it justifies bracketing the real part of the refractive index to the range 1.39–1.47 for our sensitivity tests.

As shown in Section 4.4, we estimate the ambient Hunga aerosol solution concentration to be ~50 wt%. Based on the measurements reported by Beyer et al. (1996), this corresponds to a refractive index of ~1.42 at 312 nm, which is consistent with a solution concentration of approximately 52 wt%.

To help the reader to understand our assumptions regarding the wide range of the real part of the refractive index, the following sentence has been inserted in the revised manuscript:

Pages 15–16, lines 402–425: "This wide range for the real part of the refractive index reflects the plume composition simulated by NASA Goddard Earth Observing System (GEOS) Earth system model (see Appendix F), i.e., H_2SO_4 wt% spanning roughly 40–70% (core values ~40%) and thus reasonably bracketing the ~30–80% range."

Beyer, K. D., Ravishankara, A. R., & Lovejoy, E. R. (1996). Measurements of UV refractive indices and densities of H2SO4/H2O and H2SO4/HNO3/H2O solutions. Journal of Geophysical Research: Atmospheres, 101(D9), 14519-14524.