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1 2

## Evolving Particles in the 2022 Hunga Tonga—Hunga Ha'apai Volcano Eruption Plume

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#### 16 Key Points:

- Even for the optically thin, low-latitude Tonga plume, MISR retrieved aerosol plumeheight, associated wind vectors and particle properties
   Parts of near-tropopause and mid-stratosphere aerosol layers were observed downwind over 10 days, nearly all spherical, non-light-absorbing
- The mid-stratosphere particles were smaller, but grew (ANG decreased by >20%)
   between 1/17 and 1/23, consistent with model predictions

#### 24 Abstract

25 The Multi-angle Imaging SpectroRadiometer (MISR) aboard NASA's Terra satellite observed

the Hunga Tonga—Hunga Ha'apai (HTHH) 15 January eruption plume on seven occasions

between 15 and 23 January 2022. From the MISR multi-angle, multi-spectral imagery we

retrieve aerosol plume height geometrically, along with plume-level motion vectors, and derive

radiometrically constraints on particle effective size, shape, and light-absorption properties. Parts

of two downwind aerosol layers were observed in different places and times, one concentrated in the upper troposphere (11-18 km ASL), and a mid-stratosphere layer  $\sim 23 - 30 + \text{km ASL}$ . After

the upper troposphere (11-18 km ASL), and a mid-stratosphere layer  $\sim 23 - 30 + \text{ km ASL}$ . After the initial day (1/15), the retrievals identified only spherical, non-light-absorbing particles,

typical of volcanic sulfate/water particles. The near-tropopause plume particles show constant,

medium-small (several tenths of a micron) effective size over four days. The mid-stratosphere

35 particles were consistently smaller, but retrieved effective particle size increased between 1/17

and 1/23, though they might have decreased slightly on 1/22. As a vast amount of water was also

37 injected into the stratosphere by this eruption, models predicted relatively rapid growth of sulfate

 $_{38}$  particles from the modest amounts of SO<sub>2</sub> gas injected by the eruption to high altitudes along

39 with the water (Zhu et al, 2022). MISR observations up to ten days after the eruption are

40 consistent with these model predictions. The possible decrease in stratospheric particle size after

initial growth was likely caused by evaporation, as the plume mixed with drier, ambient air.

42 Particles in the lower-elevation plume observed on 1/15 were larger than all the downwind

43 aerosols and contained significant non-spherical (likely ash) particles.

44

#### 45 Plain Language Summary

Volcanic eruptions often occur in remote locations, and typically present major hazards for 46 researchers and equipment. This creates opportunities for the frequent, global coverage satellite 47 remote sensing can make from the relative safety of space. Satellite instruments imaged the 48 massive 2022 Hunga Tonga—Hunga Ha'apai eruption plume, mapped the height of the initial 49 injections and distributions of water vapor, SO<sub>2</sub>, and aerosol particles. The Multi-angle Imaging 50 SpectroRadiomenter (MISR) instrument aboard NASA's Terra satellite offers some unique 51 capabilities in this respect, by observing the evolving particle properties in two aerosol layers, 52 one near-tropopause and one mid-stratosphere. For about a week following the initial 53 observations on 1/15, the only spherical, non-light-absorbing particles were retrieved by MISR. 54 The particles in the near-tropopause layer had relatively constant, several tenths of a micron 55 effective size. In the mid-stratosphere layer, the particles were systematically smaller, but 56 appeared to grow downwind. As a great deal of water vapor was also injected into the mid-57 stratosphere, models predicted relatively rapid growth of sulfate particles from modest amounts 58 59 of SO<sub>2</sub> injected to high altitudes along with the water. The MISR results provide observational support for these model predictions and some constraint on the associated rates, illustrating the 60 contribution satellite remote-sensing can make toward characterizing volcanic plume evolution. 61

#### 62 **1 Introduction**

The Hunga Tonga—Hunga Ha'apai (HTHH) submarine volcano in the South Pacific
(20.6° S latitude; 175.4°W longitude) began erupting on 19 December 2021 (Matoza et al.,
2022); it produced a moderate eruption at ~03:20 UTC on 14 January; a major explosive event
began at ~04:14 UTC on 15 January that propelled considerable material well into the

67 stratosphere, with some reaching into the mesosphere (Amores et al., 2022; Carr et al., 2022;

- 68 Klein 2022; Proud et al., 2022; Smart, 2022; Zuo et al., 2022). Wave perturbations were detected
- 69 globally from ground stations and satellites, within the ocean, the land surface, and through the
- atmosphere up to the ionosphere (Matoza et al., 2022; Themens et al., 2022; Wright et al., 2022).
- Relatively little  $SO_2$  and HCl were observed in the atmosphere compared to other major volcanic eruptions; for example, the 1991 Mt. Pinatubo eruption ejected ~40 times more  $SO_2$  (e.g., Carn et
- al., 2022; Millan et al. 2022; Witze, 2022). However, HTHH introduced exceptionally large
- amounts of water vapor into the stratosphere, up to unprecedented elevation (up to ~58 km) and
- in quantities not previously observed during the satellite era (Millan et al. 2022; Silletto et al.,
- <sup>76</sup> 2022). Further, significantly more sulfate particles formed and propagated in the stratosphere
- than would be expected from the relatively modest SO<sub>2</sub> injection of this eruption (Legras et al.,
- 78 2022; Taha et al., 2022).
- 79 The Multi-angle Imaging SpectroRadiometer (MISR) instrument aboard the NASA Earth
- 80 Observing System's Terra satellite observed parts of the elevated aerosol plume on at least seven
- occasions between 15 and 23 January, under fair-to-good aerosol-retrieval conditions, as the

82 plume traveled westward, traversing the Coral Sea, Australia, and the Indian Ocean (Figure 1).

83 The MISR retrievals also provide some indication of interaction between the volcanic water and

84 aerosol in the stratosphere as the plume evolved.



85

86 Figure 1. MISR Hunga Tonga-Hunga Ha'apai Volcano-plume-observation Locations, Dates, and Terra satellite Orbit numbers; the volcano is indicated with a red star. Observations of the mid-stratosphere layer, at 87  $\sim$ 23-30+ km, are shown in blue markers, those of the near-tropopause layer, at  $\sim$ 11-18 km, are shown in green. 88 The red dot indicates where non-spherical particles were retrieved at 10-14 km within a day of the main 89 90 eruption, most likely dominated by volcanic ash, and the gray dot shows where light-absorbing particles were 91 retrieved, probably smoke originating from wildfires in Australia that might have mixed with the volcanic 92 plume. All other retrievals reported spherical, non-light-absorbing particles, typical of volcanic sulfate/water 93 particles. The open circle marks a downwind plume for which MISR plume heights were retrieved, but the 94 AOD was too low for particle property retrievals.

95 The Terra satellite's near-polar orbit crosses the equator at approximately 10:30 AM local time. Each of the nine MISR cameras sweeps out a strip of imagery along the spacecraft track on 96 the day side of Earth, successively at angles ranging from  $70^{\circ}$  in the forward direction, through 97 nadir, to 70° toward the aft of the spacecraft, in each of four spectral bands centered at 446, 558, 98 672, and 866 nm (Diner et al., 1998). It takes about seven minutes for all nine cameras to view a 99 given location on Earth. The MISR swath is relatively narrow (~380 km), so sampling is 100 sporadic, especially at low latitudes where the HTHH eruption occurred. Under good, but not 101 necessarily ideal, retrieval conditions, qualitative particle microphysical properties can be 102

derived: three-to-five bins in retrieved, column-effective particle size (REPS), represented in the 103 retrieval results as the Ångström exponent (ANG) or qualitatively as "small," "medium," and 104 "large" particles between ~0.1 and ~2 microns effective radius. The exact boundaries of these 105 size bins depend on solar and viewing geometry, AOD, and particle properties, but differences in 106 REPS are much more robust, especially where the observing geometry is quite constant, as for 107 the cases in the current study (Kahn et al., 2001, Kahn and Gaitley, 2015). Under good retrieval 108 conditions, we can also retrieve two-to-four bins in column-effective particle light-absorption 109 (REPA), represented in the retrieval results as the single-scattering albedo at 558 nm wavelength 110 (SSA<sub>558</sub>), along with spherical vs. randomly oriented non-spherical particles. For the current 111 study, we used the MISR Research Aerosol retrieval algorithm (RA), which offers 1.1 km pixel 112 113 resolution, the ability to co-register multi-angle views at plume elevation, advanced radiometric calibration, and other refinements aimed at obtaining as much particle property information as 114 possible from the MISR radiances (Limbacher and Kahn, 2014; Limbacher et al., 2022). Due to 115 the stability of the instrument and the satellite, relative differences in REPS and REPA are much 116 more robust than the quantitative ANG and SSA values presented. Our conclusions in this study 117 rely primarily on relative differences in these quantities. 118

119 From a volcanological perspective, the particles below a few microns in size to which MISR particle-property retrievals are sensitive represent the lower end of ash components 120 typically emitted during eruptions. However, the MISR-retrieved aerosol optical depth (AOD) 121 captures the extinction of all particles in the column, and the very fine ash and other aerosols 122 123 within the MISR size-retrieval-sensitivity range probably capture the tail of the full ash size distribution. In themselves, these smaller components have the potential to produce far-reaching 124 effects, as they are the ones more likely to remain suspended in the atmosphere and be 125 transported long distances, impacting the radiative energy balance, air quality and health 126 downwind, as has been observed for other eruptions (e.g., Longo et al., 2010: Sahay et al., 2023; 127 Kluser et al., 2013). 128

MISR coverage is complemented by imagery from the broader-swath, single-view 129 MODerate resolution Imaging Spectroradiometer (MODIS) instrument, also aboard the Terra 130 satellite, and by layer-height curtains from space-based Cloud-Aerosol Lidar and Infrared 131 Pathfinder Satellite Observation (CALIPSO) lidar, where available. Where plume optical depth 132 is sufficient, 3-D aerosol plume-height maps can be derived geometrically from MISR multi-133 angle imagery using the MISR INteractive eXplorer (MINX) software tool, with horizontal 134 resolution of 1.1 km and uncertainty in the vertical between 250 and 500 m (Nelson et al., 2013). 135 The MISR stereo-height retrievals have been validated against contemporaneous radar and lidar 136 observations (e.g., Naud et al., 2005; Marchand et al., 2007). MINX is currently configured to 137 retrieve plume altitude values from sea level to an elevation of 30 km. The plume-height 138 139 retrievals are keyed to the layer of maximum spatial contrast in the atmospheric column; this generally identifies the optically thickest aerosol plume when multiple layers are present, unless 140 meteorological cloud or optically thick aerosol overlies other layers. As such, CALIPSO lidar 141 tends to identify thinner aerosols when they are present at higher elevations, whereas thermal 142 height-retrieval techniques tend to sample systematically lower than MISR/MINX (e.g., Flower 143 & Kahn, 2017). Aerosol plumes are rarely uniform, and MINX retrievals typically obtain a 144 range of elevations for a given plume; although the results are unlikely to capture the absolute 145 top or bottom of an aerosol layer, they often provide an indication of the plume vertical extent. 146 The associated wind vectors at plume elevation are also retrieved from MISR, based on the 147

actual movement of contrast elements in the aerosol plume itself over the seven minutes it takes

for all nine MISR cameras to image a given location. These wind vectors also allow for the

observed parallax to be corrected for plume proper motion, to produce "wind corrected" stereo

151 heights.

The constraints on particle size, shape, and light-absorption that can be derived from 152 MISR data depend upon the AOD and on the observed range of scattering angles (i.e., the angle 153 from the sun, down into the atmosphere and back up to the cameras). AOD at 558 nm (AOD<sub>558</sub>) 154 of at least 0.15 or 0.2, but also depending on surface brightness and variability, is required for 155 high-quality particle property retrievals (Kahn and Gaitley, 2015; Limbacher et al., 2022). The 156 HTHH plume data were acquired at nearly the solar equator ( $\sim 20^{\circ}$  S latitude in January). This 157 means the range of scattering angles observed by MISR was about the smallest possible, which 158 limits the sensitivity of MISR to particle properties. Nevertheless, the available MISR data do 159 yield some constraints on particle properties and their evolution during the early days of plume 160 transport, as we present here. 161

162 In this paper, we summarize the MISR results, as a contribution to the overall picture of this unique event. The instrument observed parts of the HTHH plumes with sufficient AOD to 163 obtain good retrieval results, in relatively cloud-free or broken cloud over-water areas, at least 164 seven times during the 10 days following the initial eruption. A near-source plume segment was 165 captured at an elevation of about 12 km ASL on 15 January (Figure 1, red dot), approximately 18 166 hours after the largest eruption began. On subsequent days, MISR observed parts of elevated 167 plumes toward the west, near the tropopause at  $\sim 11 - 18$  km on 19, 21, and 23 January (Figure 1, 168 green dots), and well into the stratosphere at  $\sim 23 - 30 + \text{km}$  on 17, 20, 22, and 23 January (Figure 169 1, blue dots). For 20 January, the open green dot in Figure 1 shows the location of a plume 170 segment for which the plume height was retrieved, but the AOD was too low to derive particle 171 properties. On 21 January, in addition to the near-tropopause south plume segment, an area of 172 light-absorbing particles was retrieved (grey dot in Figure 1), most likely smoke from fires in 173 northern Australia. Both upper and lower layers were observed in the same region on 23 January. 174 In Section 2, we review the MISR plume height, wind vector, and particle property retrieval 175 results. We also use data from the CALIPSO space-based lidar (Winker et al., 2010) and from 176 several AERONET surface stations (Holben et al., 1998), each acquired in the general vicinity of 177 the MISR-observed plumes, to help assess the MISR plume height and particle property results, 178 179 respectively. In Section 3, we discuss the patterns and present some inferences about plume evolution that we draw from the observations. Brief conclusions are given in Section 4. 180

#### 181 2 MISR Observations of the 2022 HTHH Eruption Plume

In this section, we summarize each MISR observation of the HTHH plume between 15 and 23 January 2022, providing our interpretation of the MINX and RA results, including comparisons with the closest AERONET and CALIOP data, where available. We focus on ANG rather than the individual retrieved components, as qualitative differences in ANG are more robust than the components and proportions that the algorithm identifies as providing a match to

- the observed top-of-atmosphere reflectances. The candidate particle optical models used for the
- 188 RA retrievals presented here are given in Table S1 in Supplemental Material.

We took several additional steps to maximize confidence in the REPS and REPA results.
 The cost function used to determine acceptable RA retrieval results is given as:

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$$\operatorname{Cost} = \frac{\sum_{\lambda} \sum_{c} \left( \frac{\sqrt{w_{\lambda,c}} * [BRF_{\lambda,c}^{\operatorname{TOA}} - BRF_{\lambda,c}^{\operatorname{mod}}]}{\operatorname{Unc}_{\lambda,c}} \right)^{2}}{\sum_{\lambda} \sum_{c} w_{\lambda,c}}$$
(1)

where BRF<sup>TOA</sup> are the MISR-observed bi-directional reflectance factors as a function of 193 wavelength ( $\lambda$ ) and camera (c). BRF<sup>mod</sup> are the model-simulated bi-directional reflectance factors 194 for a given aerosol mixture, and wavelength-dependent optical depth, surface albedo, and 195 atmospheric backscatter fraction, Unc are estimated, channel-specific radiance uncertainties, and 196 197 *w* are channel-specific weights that account for topographic shadowing, any glint contamination over water, and missing data (details are given in Limbacher & Kahn, 2019 and Limbacher et al., 198 2022). We examined the dependence of ANG and SSA results on the value of the cost function; 199 for cost function thresholds of 2 and 1 the results are similar in all cases, and the higher threshold 200 201 provides greater coverage. So, we applied a cost function upper bound of 2 to all the results presented in this paper except for the optically very thick, near-volcano 15 January case. 202

#### At low AOD, retrieval sensitivity to particle properties diminishes (e.g., Kahn and 203 Gaitley, 2015). To investigate the relationship between AOD and retrieved particle properties, 204 we plotted the dependence of ANG and SSA on the retrieved AOD. These plots are included as 205 Figures S1 and S2 in the Supplemental Material for the near-tropopause and mid-stratosphere 206 cases, respectively. Retrieved SSA is essentially independent of AOD in all cases after 15 207 January, whereas ANG tends to increase with increasing AOD<sub>558</sub>. The ANG variation appears to 208 be real, associated with geographical variation in plume properties shown in subsequent figures. 209 We set a lower bound of 0.15 on AOD<sub>558</sub> for reporting ANG and SSA statistics to exclude areas 210 where retrieval uncertainty is highest (though this affects only the 21 January case), and an upper 211 bound of 8.0 to help reduce cloud contamination. 212

- 213 Applying these criteria, we examine the seven MISR observations of the HTHH plume:
- 214

#### **Table 1**. Overview of MISR HTHH Plume Observations

Observation	Observation	Approx.	Retrieved	Retrieved	Median	Median					
date*	time	Distance from	Height	Zonal winds <sup>†</sup>	AOD <sub>558</sub>	ANG					
	[UTC]	Source [km]	[km ASL]	[m/s]							
15-Jan	~22:05	340	10 to 14	-10	~3	<~0.4					
17-Jan	~00:25	2700	27 to 30+	28	~0.7	~1.4					
19-Jan	~2:30	6500	11 to 16	24	~1.1	~0.86					
20-Jan	~2:38	7400	24 to 29	32	~0.5	~1.2					

21-Jan	~5:00	10750	23 to 26	24	~0.5	~0.94
22-Jan	~5:43	11800	23 to 26	25	~0.4	~1.3
23-Jan	~6:26	12800	15 to 17	12	~0.4	~1.0

\*19-Jan main retrieval region; 20-Jan Orbit 117501; 21-Jan South Plume †Zonal winds are approximate westward median values

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219 15 January 2022, ~22:05 UTC. MISR first imaged the HTHH plume at this time, capturing a swath centered approximately 340 km west of the volcano itself. Figure 2 shows the MISR-220 retrieved height for this plume segment at 10-14 km ASL. Careful cloud-screening in the MISR 221 RA limits the number of particle property retrievals in the MISR region over which MINX 222 223 plume heights shown in Figure 2 were derived. High-quality RA retrievals were obtained only on the west side of the study region shown in Figure 3 for this day. AOD<sub>558</sub> is ~3, the column-224 225 effective ANG is <~0.4, indicating a significant contribution from medium-large particles (i.e., >~ a micron in radius), the non-spherical AOD fraction is ~0.16, and the SSA<sub>558</sub> is ~0.99, all 226 227 suggesting the presence of some larger, non-spherical volcanic ash or ice particles (see also Table S1). Depolarization and color-ratio observations from CALIPSO also suggest the 228 229

occurrence of non-spherical ash or ice at elevations up to 40 km on this day (Sellitto et al., 2022).
 A comprehensive analysis of the 15 January 2022 plume dispersion during the first ~18 hours,

- including these MISR observations and measurements from other sources, is part of continuingwork.
- 233 On all subsequent days, discussed below, MISR retrieved only spherical, non-light-
- absorbing particles in downwind plume segments, at elevations from near the tropopause up to
- the MINX elevation retrieval limit of 30 km ASL.









- 238 Intera Oroit 11/440. (a) MODIS context image, with Wisk 45 forward view superposed (between the orange
   239 lines). The volcano location is indicated by a red star. (b) MISR MINX plume-height map, superposed on
- 239 MISR nadir-view image. (c) MISR-retrieved plume-height profile, starting point on the east side of the MISR
- field-of-view. (d) Cross-track (roughly west-east) and (e) along-track (roughly south-north) MISR-retrieved
- wind vector components, assessed over all plume points in the digitized region. (Although this plume segment
- wind vector components, assessed over an plane points in the digitized region. (Autough this plane segment was observed by MISR to the west of the volcano, the retrieved positive zonal wind indicates eastward motion.
- The zonal winds varied in elevation and time on this day, and an eastward component at the location and time
- of the MISR observation is supported by National Center for Physical Acoustics reanalysis (NCPA,
- 246 <u>https://g2s.ncpa.olemiss.edu/</u>). For all other cases in this study, the MISR-retrieved zonal wind is toward the
- 247 west.) In Panel (c), Red = zero-wind height; Blue = wind-corrected height; Green = surface elevation. No
- 248 wind-corrected heights were derived for the two lower layers at  $\sim 6$  km and  $\sim 11$  km elevation in Panel c, as the
- origin of digitization was selected for the more prominent layer at ~ 12 km.



Figure 3. MISR Research Algorithm particle property retrieval results for 15 January 2022, Terra Orbit
117440. (a) MISR context image with study region outlined; the relationship of this region to the HTHH
volcano is shown in Figure 2a. (b) AOD<sub>558</sub> in the less cloudy parts of the study region; (c) ANG assessed
between 446 and 866 nm; (d) SSA<sub>558</sub>; (e) Non-spherical AOD<sub>558</sub> fraction. No cost-function filter was applied
in this one case, but to improve confidence in the particle property retrieval results, ANG and SSA were
assessed over the AOD range 0.15 < AOD<sub>558</sub> <8.0. Digital data for the MISR RA results are given in Table S1.</li>

17 January 2022, ~00:25 UTC. On this day, MISR captured a part of the HTHH plume off the 257 northeast coast of Australia, ~2,700 km west of the volcano, and ~450 km north of the Lucinda 258 259 AERONET site. Plume heights reached from 27 and 30+ km ASL (Figure 4). (As 30 km is the highest elevation allowed by the MINX algorithm, some contrast elements might have occurred 260 above the MINX-reported peak altitude.) AOD<sub>558</sub> had dropped from the previous MISR plume 261 observation to ~0.7, with ANG ~1.4 (Figure 5 and Table S1). Such small particles provided 262 enough opacity in the MISR blue band (at 446 nm) for contrast features to allow a plume-height 263 retrieval for the 30+ km layer (Figure 4c). However, the elevated plume particles were 264 apparently too small and optically thin in the MISR red band (at 672 nm) to produce sufficient 265 contrast, and only a near-surface layer and a weaker layer around 12 km were retrieved in this 266 channel (Figures 4b). CALIOP lidar confirms the presence of the ~30 km layer in this general 267

area (Figure 4d). The AERONET station at Lucinda obtained contemporaneous measurements

- of AOD<sub>550</sub>  $\sim$ 0.7 and ANG  $\sim$ 1.55 (Figures 4f and 4g), confirming to the extent possible the
- 270 presence of an aerosol layer similar to that retrieved by MISR.



Figure 4. MISR MINX Plume-height retrieval results for 17 January 2022, Terra Orbit 117456, 272 ~00:25 UTC, acquired north of Queensland, Australia, along with Lucinda AERONET ground-273 station retrievals. (a) MODIS context image, with MISR 60° forward view superposed (between 274 the orange lines). The MISR retrieval region is indicated by the yellow arrow. The location of the 275 Lucinda AERONET station on the northeast Australia coast, marked with a yellow asterisk 276 within the MISR swath, ~450 km south of the over-ocean region where the MISR retrievals were 277 278 performed, was still apparently within the plume. (For scale, the MISR swath is roughly 380 km wide.) (b) Red-band MISR-MINX plume-height histogram, showing a weak layer near the 279 tropopause at 12-14 km and another near-surface. (c) Blue-band MISR plume-height histogram, 280 showing a layer at 27-30+ km. (d) CALIOP Level 1, version 4.51, total attenuated backscatter 281 (km<sup>-1</sup> sr<sup>-1</sup>) at 532 nm, acquired at ~05:28 UTC on 17 January, roughly 5 hours after and 800 km 282 downwind of the MISR observation at the nearest point. (The inset gives the orbit location, and 283 the red arrow highlights the likely plume layer.) (e) Lucinda AERONET AOD for 24 January 284 2022, showing the AOD peak on 17 January. (f) Lucinda AERONET AOD for 17 January 2022. 285 (g) Lucinda AERONET Ångström Exponent for 17 January 2022. (AERONET data were 286 provided by Thomas Schroeder and the AERONET team.) 287



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**Figure 5**. MISR RA particle property retrieval results, same as Figure 3, but for 17 January 2022, Terra Orbit 117456. Note that the scale for panel (e) is logarithmic here to show differences, given the range of in values.

*19 January 2022, ~2:30 UTC.* MISR observed a plume segment southwest of Java (Figure S3 in
Supplemental Material), yielding a plume height of 11-16 km and MINX zonal winds at plume
elevation in excess of 24 m/s toward the west. Neither CALIPSO nor ICESat-2 have
measurements sufficiently proximal to the MISR observations in space and time to provide
validation of the MISR height retrievals. At these low latitudes, the MISR layer would be below,
but in the vicinity of, the tropopause elevation. In the least cloud-affected part of the field-ofview, the MISR RA retrieved AOD<sub>558</sub> ~1.1, ANG ~0.8, with spherical, non-light-absorbing

298 particles dominating (Figure S4 and Table S1).

299 20 January 2022, ~2:38 UTC. On 20 January MISR observed part of the HTHH plume along the west coast of Australia, about 7,400 km west of the volcano. Here the plume was elevated 300 between 24 and 29 km ASL, with zonal winds of ~30 m/s (Figure S5). CALIOP identified a 301 plume at  $\sim 25 - 27$  km ASL about 7 hours earlier and 400 km downwind of the MISR 302 observation. MISR median AOD<sub>558</sub> was about 0.5, most ANG values ~ 1.1, and the particles 303 were again spherical and non-light-absorbing (Figure S6 and Table S1). For comparison, the 304 Learmonth AERONET, about 350 km northeast of the MISR study region along the Australia 305 west coast, obtained AOD<sub>500</sub> and ANG at the closest observations to MISR overpass time of 0.7 306 and 1.05, respectively (Figures S6g and S6h). The somewhat larger AOD at the AERONET site 307 could be due to heterogeneity in the aerosol plume after a 5-day transit, with possible 308

309 contributions from continental aerosol from Australian sources. There was a second MISR

overpass of a part of the likely volcanic plume ~2,700 km further west, at ~4:14 UTC. The

MISR-MINX retrieval obtained a plume height of 13-17 km ASL, with the hint of another layer 125 28 km ASL (Figure S7) because the AOD must be been to device magning following the test of the second seco

at 25-28 km ASL (Figure S7); however, the AOD was too low to derive meaningful particle

313 microphysical properties from the data.

21 January 2022, ~5:00 UTC. The plume observed by MISR on this day was in the central 314 Indian Ocean, about 10,750 km from the source. MISR retrieved a consistent plume elevation of 315 23-26 km ASL (Figure S8). (ICESat-2 indicated several lower-level aerosol layers between 8 and 316 317 12 km, but no retrievals are currently available that sample above 14 km.) The MISR study region on this day contained two plume segments, north and south, that showed distinctly 318 different particle microphysical properties. As with most of the other downwind plume parts 319 captured by MISR, the South segment on 21 January contained small-medium, spherical, non-320 light absorbing particles, with AOD<sub>558</sub> ~0.5, ANG ~0.95-1.0, and SSA<sub>558</sub> ~1.0 (Figure S9 and 321 Table S1). 322

However, for the North segment, AOD<sub>558</sub> is ~0.9, ANG is ~1.4-1.5, and SSA<sub>558</sub> is 323 324 between 0.94 and 0.96 (Figure S10 and Table S1). The low SSA<sub>558</sub> and smaller effective particle size suggest the North segment might contain smoke. This seems likely, as major fires were 325 burning in northeast and northwest Australia, producing extensive smoke plumes visible in 326 MODIS imagery from 16 to 18 January (https://worldview.earthdata.nasa.gov; last accessed 01 327 May 2023). Trajectory analysis from NOAA's HySPLIT model (Rolph et al., 2017) indicates 328 that transport from across north Australia to the location of the MISR plume observation on 21 329 January (~21.4°S, 77.3°E) occurred in 3-4 days at elevations from 18 to over 27 km. This result 330 also represents a demonstration of our ability to derive distinct aerosol type differences from the 331 MISR multi-angle, multi-spectral data. 332

333 22 January 2022, ~5:43 UTC. MISR observed a plume segment at 23-26 km ASL on 22

January, about 11,800 km from the source (Figure S11). Yet, the MISR RA derived a mean

AOD<sub>558</sub> around 0.4, within the range for which particle properties can be retrieved over dark water (Figure S12 and Table S1). The particles are all of small size (ANG  $\sim$ 1.3) and are still

spherical and non-light absorbing, as for all previous downwind retrievals associated with the

338 volcano plume.

23 January 2022, ~6:26 UTC. This was the final occasion on which MISR captured a segment 339 of the HTHH eruption plume having sufficient definition to report plume elevations with some 340 341 confidence. In this case, we derive particle microphysical properties qualitatively in two distinct layers. By the time of these observations, the plume particles had reached the vicinity of Réunion 342 Island, ~12,800 km west of the volcano and ~720 km east of Madagascar. The lower layer was 343 concentrated 15-17 km ASL, whereas the thinner upper layer concentrated at ~24-26 km above 344 the ocean surface, extending over the lower layer as well as to the northwest and southeast of the 345 lower layer (Figure S13b, c). The MISR retrieval indicates westward motion at ~ 8-12 m/s 346 (Figure S13e, f). For the "mixed-height" layer, dominated by the lower, optically thicker layer, 347 total-column AOD<sub>558</sub> was retrieved at ~ 0.4 and ANG ~1.0 (Figure S14 and Table S1). For the 348 elevated layer alone, AOD<sub>558</sub> was <~ 0.3 and ANG ~1.1. The AERONET site at Réunion, just 349

north of the MISR retrieval region, reported AOD500 ~0.4 and ANG ~0.63 at the 6:26 UTC
 MISR overpass time, i.e., larger particles than those observed by MISR (Figure S13).

There was also a stratospheric balloon flight deployed from La Réunion island itself 352  $(21.1^{\circ} \text{ S}; 55.3^{\circ} \text{ E})$  on this day (20:04 - 21:35 UTC), carrying an optical counter with sensitivity 353 to particle sizes from 0.2  $\mu$ m to ~30 mm diameter in 19 bins (Kloss et al., 2022). They also had 354 a ground-based backscatter lidar. The balloon identified plume layers at 22.6 and 24.9 km ASL, 355 the upper layer corresponding to the upper layer identified by MISR (Fig. S13c, d). The lower 356 layer identified by MISR is about 6 km below the lower layer reported by the *in situ* 357 358 measurements. (There are also discrepancies in layer elevations between the balloon and the ground-based lidar, likely due to differences in spatial sampling combined with layer 359 heterogeneity.) Consistent with the MISR results, the optical counter found spherical, non-light 360 absorbing particles with radii <0.5 µm in the upper layer. In their 22.6 km layer, they report 361 some light-absorbing particles, which could be transported smoke, as we observed with MISR on 362 21 January. 363

#### 364 **3 Discussion – MISR Constraints on the Evolution of the HTHH Aerosol Plume**

365 The altitude reached by volcanic emissions depends on ejection force, plume buoyancy, atmospheric stability profile, wind shear, and particle properties. Generally, ash particles are 366 likely to settle more rapidly than smaller, lighter, aerosol components, especially those that tend 367 to form after eruption from water vapor and other gases, such as typical sulfate/water particles in 368 volcanic plumes. On 15 January, MISR retrieved particles containing a significant fraction of 369 non-spherical components, with ANG <~0.4, indicating medium-large particles (Table S1), most 370 likely dominated by volcanic ash. A complimentary study by Sellitto et al. (2022) concluded that 371 ash was deposited rapidly between 19:00 UTC on 15 January and 02:00 UTC on 16 January, 372 based on frequent observations made with the geostationary Himawari 8 satellite. The MISR 373 observation, obtained at 22:05 UTC on 15 January, falls within this window. As such, the MISR 374 observation was likely acquired during a period of rapid ash deposition. MISR ash detection on 375 this day is further supported by the lack of ash markers identified in subsequent MISR retrievals; 376 377 between 17 and 23 January MISR retrievals consistently found spherical, non-light-absorbing aerosols, more likely composed of sulfate/water than ash. (The other exception occurred on 21 378 January, when in addition to the volcanic plume, MISR observed what was likely transported 379 smoke (Figure S10).) We note that Colombier et al. (2023) posit non-spherical sea salt particles 380 might be part of the 15 January atmospheric aerosol load, given the submarine nature of the 381 eruption and the presence of sea salt in surface samples acquired on nearby islands. With MISR 382 we cannot distinguish such particles from ash, so our data have nothing to contribute about this 383 possibility. 384

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Other satellite observations identified an SO<sub>2</sub> gas plume ejected during the HTHH 386 eruption and tracked the resulting sulfate particles to altitudes exceeding 45 km (Sellitto et al., 387 2022; Taha et al., 2022), possibly up to where the plume achieved neutral buoyancy and/or 388 where ambient conditions, such as the anomalously high concentration of water vapor, favored 389 rapid sulfate formation through oxidation and hydration processes. The apparent vertical 390 separation of sulfate/water and ash plumes has also been observed in other volcanic eruptions, 391 such as Grimsvotn in 2011 (Flower & Kahn, 2020). Preferential settling of ash likely contributed 392 to the lack of larger, non-spherical particles observed further downwind from the volcano after 393

15 January. The observation of localized ash-fall around the volcano immediately following the
 eruption (e.g., *Thompson*, 2022) also supports the rapid-ash-deposition hypothesis.

Two aerosol layers were observed in the downwind MISR plume detections, one near-397 tropopause and one mid-stratosphere. For the near-tropopause layer, AOD<sub>558</sub> exceeded unity on 398 19 January, diminishing to ~0.47 on 21 January (South plume) ~0.4 on 22 January, and 0.26 by 399 23 January in the MISR retrievals (Table S1). These retrievals indicate spherical, non-light-400 absorbing particles, with median ANG between 0.85 and about 1.0, i.e., small-medium particles. 401 possibly decreasing very gradually in effective size (Figure 6). With the sparse MISR sampling 402 of these extended aerosol layers, plume heterogeneity is likely, so the values reported should be 403 404 considered qualitatively representative of overall plume properties during transport, with higher confidence in the relative changes than the absolute values. The North plume segment on 21 405 January has distinctly different microphysical properties than the adjacent South plume segment 406 observed at the same elevation: significantly higher median AOD<sub>558</sub> (~0.9), lower SSA<sub>558</sub> 407 (~0.96), and larger ANG (~1.5). The North plume properties are typical of transported wildfire 408 smoke, and model trajectories from burning regions in parts of north Australia support this 409 suggestion (see Section 2 above). These results also contribute to validating the qualitative MISR 410 particle microphysical property retrievals, as they show distinctions that are at least consistent 411





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**Figure 6.** MISR-retrieved Ångström exponent (ANG, inversely proportional to effective particle size) for the MISR observations of the near-source HTHH plume on 15 January (red dot), and for the near-tropopause (green dots) and stratosphere (blue dots and gray dot) plumes downwind. Observation dates and plume heights are given in the annotations.

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AOD<sub>558</sub> in the mid-stratosphere layer diminished systematically from about 0.7 to  $\sim 0.26$ 419 between 17 and 23 January (Table S1). Essentially spherical, non-light-absorbing particles were 420 retrieved in all cases for this layer. Of particular interest is the effective particle size, which is 421 inversely proportional to the MISR-retrieved ANG plotted for the available cases in Figure 6. 422 For the plume segments in the mid-stratosphere layer (blue dots), the effective particle size was 423 always smaller than that for the near-tropopause layer (green dots), and the retrieved effective 424 particle size (REPS) actually increased between 17 and 20 January, then appears to have 425 decreased by 22 January and possibly increased again in the lowest-AOD observation on 23 426 January. From the available snapshots, particle evolution can only be inferred; if these 427 428 observations capture the evolution of particles in the layer near 30 km, either coagulation of particles in the layer or the condensation of water might be responsible for the observed particle 429 growth. The layer mid-visible optical depth was ~0.4 on 22 January (Table S1) and the layer 430

vertical extent was at least 5 km (Figure S11c), so particle concentrations were fairly low, which 431 does not favour particle collision and coagulation. However, an unprecedented amount of water 432 vapor was present at these elevations; ordinarily, the stratosphere at ~30 km is very dry, whereas 433 HTHH was a "wet" eruption, and Microwave Limb Sounder (MLS) data show that significant 434 water was injected to these elevations (Millan et al., 2022). This supports an explanation for the 435 apparent particle growth as the result of particle hydration, either by adsorption or by ice 436 deposition. Ice deposition on volcanic particles is a well-known phenomenon in wet (i.e., 437 phreatomagmatic) eruption plumes (e.g., Van Eaton et al., 2012), an example of the classic 438

439 juxtaposition of fire and ice.

#### 440 **4 Conclusions**

As the MISR swath-width is relatively narrow, inferences drawn about HTHH plume 441 evolution from the snapshots of this low-latitude eruption presented here must be considered in 442 443 the context of broader-swath observation provided by other instruments, as cited above. Yet, MISR does contribute unique information, particularly about changes in height-resolved particle 444 properties during the week following the eruption. The MISR-retrieved spectral AOD results 445 generally align with those obtained by ground-based sun photometers, which provide some 446 validation for the MISR aerosol amount (AOD) and particle size (ANG) retrievals. Similarly, 447 three cases where the CALIPSO lidar observations of plume height were sufficiently close in 448 space and time to the MISR observations support the MISR-MINX plume-height results. 449

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451 MISR captured two distinct layers of the HTHH eruption plume downwind, one neartropopause and one mid-stratosphere. On 15 January, a plume rich in medium-large, non-452 spherical particles (likely volcanic ash) was observed a few hundred kilometers to the west of the 453 volcano. Thereafter, only small or small-medium, spherical, non-light-absorbing particles, 454 typical of volcanic sulfate/water, were retrieved by MISR. By 23 January, the sulfate-like HTHH 455 aerosol plume had remained suspended for more than a week, had travelled at least a third of the 456 way around the globe, and was still sufficiently optically thick to support MISR plume-height 457 and particle-property retrievals, though with somewhat lower confidence than the higher-AOD 458 459 cases. 460

Particles in the mid-stratosphere layer (blue dots in Figures 1 and 6) were systematically 461 smaller than the near-tropopause particles (green dots). The mid-stratosphere particles apparently 462 grew in size between 17 and 20 January, likely due to hydration or water condensation, then 463 seem to have fluctuated in size between 22 and 23 January, possibly due to some combination of 464 measurements sampling different parts of a heterogeneous plume and evaporation as the plume 465 mixed with drier air and dissipated downwind. For the near-tropopause layer, particle size 466 remained relatively constant or very gradually diminished between 19 and 23 January. Due to the 467 stability of the instrument and the satellite, these relative size differences, retrieved from MISR, 468 are much more robust than absolute values. As such, our main conclusions rely on relative 469 differences in particle microphysical properties and showconsistency with model predictions 470 (e.g., Zhu et al., 2022). 471

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We note also that after we originally submitted the current paper, several preliminary
studies appeared relevant to key aspects of our conclusions. Specifically, Biochu et al. (2024)
combined HIMAWARI-8 geostationary thermal infrared imagery with CALIPSO lidar to

- identify ash on 15 January, and subsequently smaller, but initially rapidly growing, stratospheric
- aerosol with estimated 0.3-0.5  $\mu$ m radius, which they interpret as sulfate and track for up to 1.4
- 478 years thereafter. They support their satellite-data interpretation with measurements from multiple
- 479 low-latitude AERONET stations, including the Lucinda and Learmonth stations that we use for
- validation in the current study. Gupta et al. (2024) used Stratospheric Aerosol and Gas
- 481 Experiment-III (SAGE-III) observations to characterize stratospheric aerosol amount and
- extinction associated with the HTHH eruption; they highlight the importance of knowing the sulfate aerosol amount and properties, showing that although water vapor added to the
- sulfate aerosol amount and properties, showing that although water vapor added to the
   stratosphere would produce surface warming, this would be offset by a net cooling due to
- stratosphere would produce surface warning, this would be onset by a net c
   stratospheric ozone loss and sulfate aerosol production.
- 486

The MISR results presented here demonstrate our space-based multi-angle, multi-spectral aerosol retrieval capabilities, and represent the MISR contribution to ongoing investigations by the wider community aimed at deriving as much as possible about HTHH plume-particle evolution from satellite remote-sensing data and modelling.

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499

## 500 **Open Research**

MISR data are freely available for download from NASA's MISR data repository 501 (https://l0dup05.larc.nasa.gov/MISR/cgi-bin/MISR/main.cgi), maintained by the NASA Langley 502 Research Center (LaRC) Atmospheric Science Data Center (ASDC). The MISR INteractive 503 eXplorer (MINX) program, for determining plume altitude and associated motion vertors, is a 504 stand-alone software package developed at the NASA Jet Propulsion Laboratory (JPL) and 505 distributed through Github (https://github.com/nasa/MINX/releases). MODIS true color imagery 506 are accessed through the NASA Worldview application (https://worldview.earthdata.nasa.gov), 507 part of the NASA Earth Observing System Data and Information System (EOSDIS). The 508 509 AERONET and CALIPSO data are freely available from their respective archives at https://aeronet.gsfc.nasa.gov and https://subset.larc.nasa.gov/calipso/. MISR RA and MINX 510 results generated in this work will be accessed through the NASA Langley ASDC, where the 511 MISR Standard Products are also archived (https://eosweb.larc.nasa.gov/project/MISR). All the 512 results of the study are presented in the paper itself and supplemental material, including the key 513 data statistics associated with this study. 514

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#### 679 Figure Captions

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Figure 1. MISR Hunga Tonga—Hunga Ha'apai Volcano-plume-observation Locations, Dates, 681 and Terra satellite Orbit numbers; the volcano is indicated with a red star. Observations of the 682 mid-stratosphere layer, at  $\sim 23-30+$  km, are shown in blue markers, those of the near-tropopause 683 layer, at ~11-18 km, are shown in green. The red dot shows where non-spherical particles were 684 retrieved at 10-14 km within a day of the main eruption, most likely dominated by volcanic ash, 685 and the gray dot shows where light-absorbing particles were retrieved, probably smoke 686 originating from wildfires in Australia that might have mixed with the volcanic plume. All other 687 retrievals reported spherical, non-light-absorbing particles, typical of volcanic sulfate/water 688 particles. The open circle marks a downwind plume for which MISR plume heights were 689 retrieved, but the AOD was too low for particle property retrievals. 690

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692 Figure 2. MISR MINX Plume-height and Wind Vector retrieval results for 15 January 2022 at ~22:05 UTC, Terra Orbit 117440. (a) MODIS context image, with MISR 45° forward view 693 superposed (between the orange lines). The volcano location is indicated by a red star. (b) MISR 694 MINX plume-height map, superposed on MISR nadir-view image. (c) MISR-retrieved plume-695 height profile, starting point on the east side of the MISR field-of-view. (d) Cross-track (roughly 696 west-east) and (e) along-track (roughly south-north) MISR-retrieved wind vector components, 697 698 assessed over all plume points in the digitized region. (Although this plume segment was observed by MISR to the west of the volcano, the retrieved positive zonal wind indicates 699 eastward motion. The zonal winds varied in elevation and time on this day, and an eastward 700 component at the location and time of the MISR observation is supported by National Center for 701 Physical Acoustics reanalysis (NCPA, https://g2s.ncpa.olemiss.edu/). For all other cases in this 702 study, the MISR-retrieved zonal wind is toward the west.) In Panel (c), Red = zero-wind height; 703 Blue = wind-corrected height; Green = surface elevation. No wind-corrected heights were 704 derived for the two lower layers at ~6 km and ~11 km elevation in Panel c, as the origin of 705 digitization was selected for the more prominent layer at ~ 12 km. 706

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Figure 3. MISR Research Algorithm particle property retrieval results for 15 January 2022,
Terra Orbit 117440. (a) MISR context image with study region outlined; the relationship of this
region to the HTHH volcano is shown in Figure 2a. (b) AOD at 558 nm in the less cloudy parts
of the study region; (c) ANG assessed between 446 and 866 nm; (d) SSA at 558 nm; (e) Nonspherical AOD<sub>558</sub> fraction. No cost-function filter was applied in this one case, but to improve
confidence in the particle property retrieval results, ANG and SSA were assessed over the AOD

- range  $0.15 < AOD_{558} < 8.0$ . Digital data for the MISR RA results are given in Table S1.
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**Figure 4.** MISR MINX Plume-height retrieval results for 17 January 2022, Terra Orbit 117456, ~00:25 UTC, acquired north of Queensland, Australia, along with Lucinda AERONET ground-

~00:25 UTC, acquired north of Queensland, Australia, along with Lucinda AERONET groundstation retrievals. (a) MODIS context image, with MISR 60° forward view superposed (between

the orange lines). The MISR retrieval region is indicated by the yellow arrow. The location of the

Lucinda AERONET station on the northeast Australia coast, marked with a yellow asterisk

within the MISR swath, ~450 km south of the over-ocean region where the MISR retrievals were

performed, was still apparently within the plume. (For scale, the MISR swath is roughly 380 km

wide.) (b) Red-band MISR-MINX plume-height histogram, showing a weak layer near the

tropopause at 12-14 km and another near-surface. (c) Blue-band MISR plume-height histogram,

- showing a layer at 27-30+ km. (d) CALIOP Level 1, version 4.51, total attenuated backscatter
- $(km^{-1} sr^{-1})$  at 532 nm, acquired at ~05:28 UTC on 17 January, roughly 5 hours after and 800 km
- downwind of the MISR observation at the nearest point. (The inset gives the orbit location, and
- the red arrow highlights the likely plume layer.) (e) Lucinda AERONET AOD for 24 January
- 2022, showing the AOD peak on 17 January. (f) Lucinda AERONET AOD for 17 January 2022.
- (g) Lucinda AERONET Ångström Exponent for 17 January 2022. (AERONET data were
- 731 provided by Thomas Schroeder and the AERONET team.)
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- **Figure 5**. MISR RA particle property retrieval results, same as Figure 3, but for 17 January
- 7342022, Terra Orbit 117456. Note that the scale for panel (e) is logarithmic here to show
- differences, given the range of in values.
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- **Figure 6.** MISR-retrieved Ångström exponent (ANG, inversely proportional to effective particle
- size) for the MISR observations of the near-source HTHH plume on 15 January (red dot), and for
- the near-tropopause (green dots) and stratosphere (blue dots and gray dot) plumes downwind.
- 740 Observation dates and plume heights are given in the annotations.
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