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# 1 **Global perturbation of stratospheric water and aerosol burden by Hunga eruption**

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31

32

33        **Abstract**

34        The eruption of the submarine Hunga volcano in January 2022 was associated with a powerful  
35 blast that injected volcanic material to altitudes up to 58 km. From a combination of various types  
36 of satellite and ground-based observations supported by transport modeling, we show evidence for  
37 an unprecedented increase in the global stratospheric water mass by 13% as compared to  
38 climatological levels, and a 5-fold increase of stratospheric aerosol load, the highest in the last  
39 three decades. Owing to the extreme injection altitude, the volcanic plume has circumnavigated  
40 the Earth in only one week and dispersed nearly pole-to-pole in three months. The unique nature  
41 and magnitude of the global stratospheric perturbation by the Hunga eruption ranks it among the  
42 most remarkable climatic events in the modern observation era, with a range of potential persistent  
43 repercussions for stratospheric composition and climate.

44        **Introduction**

45        The main eruption of the Hunga submarine volcano (Tonga, 20.54°S, 175.38°W) on 15  
46 January 2022 was likely the most explosive event of the modern observational era, with an  
47 estimated Volcanic Explosivity Index (VEI) of 5.8 (Poli and Shapiro, 2022). In the historical  
48 record, the Lamb wave triggered by the initial explosion is only comparable to that of the eruption  
49 of Mount Krakatoa in 1883 (Matoza et al., 2022; Wright et al., 2022). Stereoscopic analysis of  
50 geostationary satellite images shows that the volcanic plume reached up to about 58 km (Carr et  
51 al., 2022), resulting in the direct injection of volcanic gases and vaporised seawater from the  
52 magmatic chamber together with tropospheric moisture entrained by the eruptive updraft.

53        The dryness of the stratosphere is largely conditioned by the transit of the air masses through  
54 the cold tropical tropopause where freeze-drying usually limits the amount of water entering the  
55 stratosphere to a few ppmv (Brewer, 1949; Mote et al., 1996; Bonazzola and Haynes, 2004). As  
56 the atmospheric radiation budget is particularly sensitive to water vapour changes in the upper  
57 troposphere and lower stratosphere (e.g., Forster and Shine, 2002; Riese et al., 2012), even small  
58 changes in the stratospheric water content can lead to significant radiative forcing (Solomon et al.,  
59 2010) and alter stratospheric ozone chemistry (Anderson et al., 2012). The increase in  
60 stratospheric water vapour concentrations by a few ppmv simulated by current chemistry climate  
61 models in response to global warming may cause substantial positive climate feedbacks amplifying  
62 surface warming (Dessler et al., 2013). A rise in stratospheric water vapour also induces significant

63 changes in atmospheric circulation, increasing the poleward and upward shift of subtropical jet  
64 streams and intensifying the stratospheric Brewer-Dobson circulation by about 30% (Li and  
65 Newman, 2020), with further potential implications for surface climate.

66 Early studies of volcanic columns (e.g., Glaze et al., 1997) advocated that deep volcanic plumes,  
67 such as those of 1815 Tambora or 1883 Krakatoa, may have led to significant stratospheric  
68 hydration. Water vapor constitutes about 80% in volume of the erupted gas (Holland, 1978; Pinto  
69 et al., 1989) and a few percent of the total mass of ejected material which, for Hunga, ranges from  
70 2,900 Tg (Yuen et al., 2022) to 13,000 Tg (Poli and Shapiro, 2022). Additional moisture may also  
71 be entrained from the troposphere (Glaze et al., 1997; Joshi and Jones, 2009). However, this  
72 stratospheric moistening conjecture had never been proven from observations. Due to  
73 condensation near the cold point tropopause, moderately explosive eruptions of the last two  
74 decades only generated limited water vapour injections, in contrast with their substantial impacts  
75 on the stratospheric sulfur and aerosol budget (Sioris et al., 2015; 2016). Pitari and Mancini (2002)  
76 proposed that the 1991 eruption of Mount Pinatubo injected about 37 Tg of water but this estimate  
77 was based solely on modeling considerations. The Hunga eruption on January 15, 2022 provides  
78 observational evidence for significant stratospheric hydration after a major volcanic eruption and  
79 recent studies (Millan et al., 2022; Xu et al., 2022) estimated that Hunga injected about 139 to 146  
80 Tg of water into the stratosphere.

81 In this paper, we describe and quantify the stratospheric repercussions of the unique natural  
82 experiment in the middle atmosphere provided by the Hunga eruption. We investigate the  
83 formation and evolution of the stratospheric moisture and sulfate aerosol plume at a wide range of  
84 scales - from minutes and kilometres to monthly and planetary scales - using a synergy of satellite  
85 and ground-based observations supported by transport modeling. Given the outstanding magnitude  
86 of stratospheric moistening, and the absence of efficient sinks of moisture in the stratosphere, the  
87 Hunga eruption can be said to have initiated a new era in stratospheric gaseous chemistry and  
88 particle microphysics with a wide range of potential long-lasting repercussions for the global  
89 stratospheric composition and dynamics.

90

91

## 92 Volcanic injection into the middle atmosphere

93 While the Hunga eruptive sequence on January 15 (D+0) started around 04:05 UTC  
94 (Astafyeva et al., 2022), the paroxysmal blast occurred at 04:16 UTC (Poli and Shapiro, 2022;  
95 Matoza et al., 2022). At 04:25, the main volcanic plume reached its top recorded altitude of 58 km  
96 (Fig. 1A) with an ascent speed of at least 40 m/s over the previous 10 minutes, as shown by  
97 stereoscopic analysis of high-resolution imaging by GOES-17 and Himawari-8 geostationary  
98 satellites (Fig. S1A, Supplementary notes, Movie S1). The time evolution of the cloud top height  
99 reveals a complex eruption scenario of successive updrafts, in agreement with observations of  
100 infrasound waves suggesting three emission events (Podglajen et al., 2022). Numerical simulations  
101 of large eruptive columns (e.g. Woods, 1988) show that their temperature exceeds that of ambient  
102 air by hundreds of K at the tropopause. Thus, the initial plume has effectively bypassed the cold  
103 trap of the tropical tropopause and lower stratosphere.

104 The stratospheric umbrella cloud expanded at an altitude nearing 40 km within the first hour  
105 of the eruption to cover 150,000 to 200,000 km<sup>2</sup> (Fig. S1B). The dome of the ice cloud above 40  
106 km, exposed to higher temperatures in the upper stratosphere (Fig. 1B), has entirely sublimated  
107 within an hour after the first blast (Fig. 1C). The lower umbrella, topping at around 35 km altitude,  
108 persisted longer and, carried by fast stratospheric easterlies, expanded 500 km to the West (Fig.  
109 1A) and grew to 320,000 km<sup>2</sup> in three hours, nearly the size of Germany (Fig. S1B).

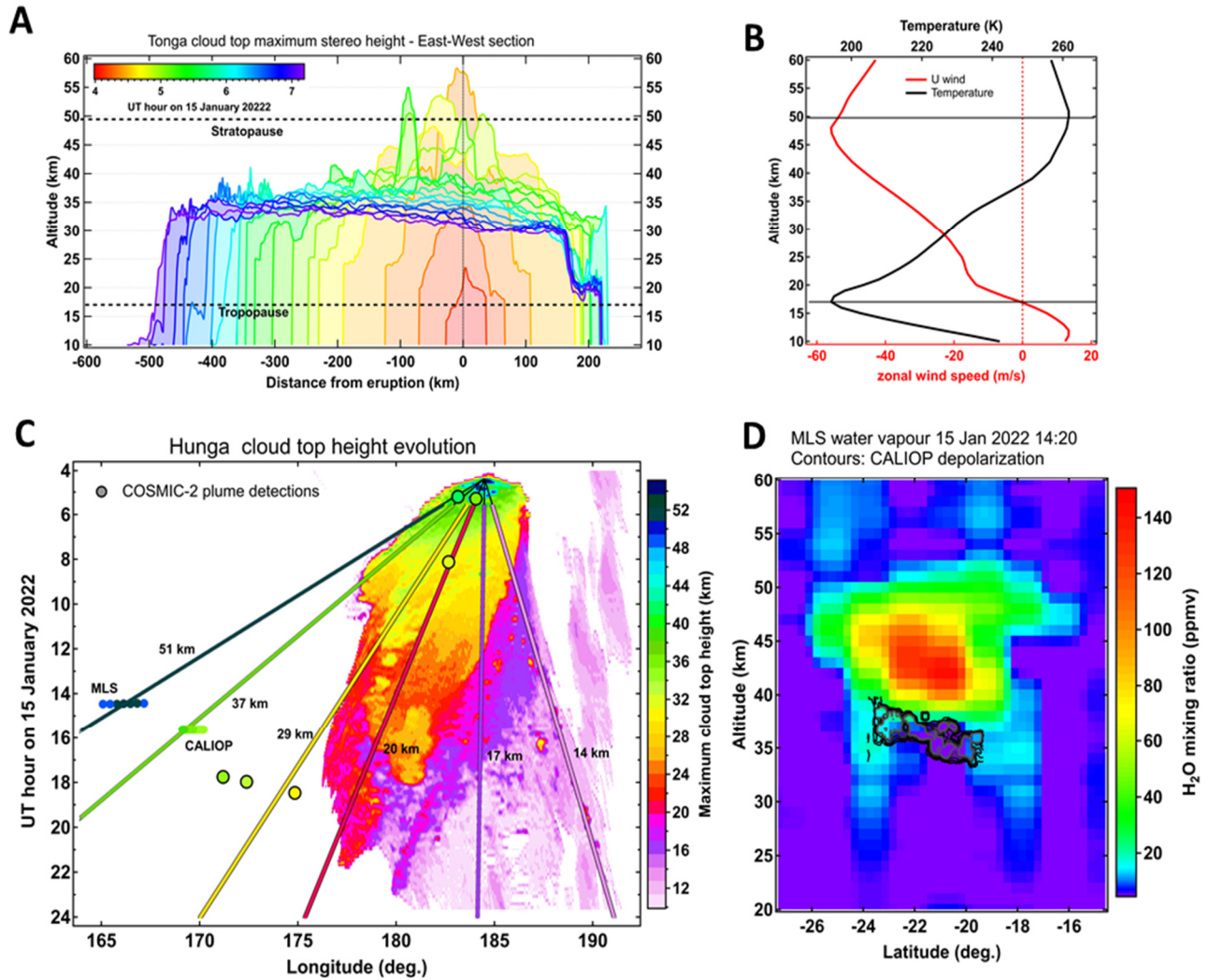
110 The formation and persistence of ice at such high altitude, although foreseen by modelling  
111 studies (Glaze et al., 1997; Textor et al., 2003), implies near ice-saturation and humidities more  
112 than three orders of magnitude larger than typically encountered in the stratosphere. Such near-  
113 saturation stratospheric water vapour in the plume is confirmed by Global Navigation Satellite  
114 System (GNSS) COSMIC-2 radio occultation soundings (hereafter GNSS-RO) on D0 downwind  
115 of the eruption (marked as circles in Fig. 1C). Extreme anomalies in bending angles and refractivity  
116 at altitudes of 30 to 40 km for about an hour after the eruption translate into strikingly-high  
117 stratospheric water vapour anomalies up to 15,000 ppmv at 37 km (Fig. S2B). Near saturation  
118 conditions are also revealed in later soundings and even persisted for 3 - 4 days at 20 - 30 km (Fig.  
119 S3).

120 In the case of a very fast injection such as that of Hunga, the main factors influencing the  
121 amount of water remaining in the stratosphere and hence the scavenging of volcanic gases by  
122 sedimenting ice appear to be the background temperature profile, plume top altitude and horizontal

123 extent of the umbrella cloud. Extrapolating the two early GNSS-RO profiles to the whole area of  
124 the young umbrella cloud and neglecting the remaining ice leads to a stratospheric total water  
125 injection lying between 100 - 150 Tg, similar to what would be obtained assuming a saturated  
126 stratospheric column up to ~33 km. Comparison with later estimates of the mass of injected water  
127 from Microwave Limb Sounder (MLS) observations showing 119 - 137 Tg (Fig. S6,  
128 Supplementary notes) further suggests that the ice which survived fall-out and later sublimated  
129 only plays a marginal role in the injection budget. The volume of water injected into the  
130 stratosphere corresponds to the average amount of water discharged by the Amazon river  
131 ( $2.09 \times 10^5 \text{ m}^3/\text{s}$ ) over about 10 minutes. Note that the peak volumetric discharge of the volcanic  
132 plume was estimated at  $\sim 9 \times 10^5 \text{ m}^3/\text{s}$  (Yuen et al., 2022).

133 The motion and lifetime of volcanic ice clouds at different heights (Fig. 1C) can be explained  
134 by the easterly-sheared background flow and sublimation due to dilution within a warmer and drier  
135 environment (Fig. 1B). At higher levels, the plume was subject to faster westward advection, but  
136 also lower environmental humidity/higher temperatures towards the stratopause leading to a  
137 quicker sublimation (within an hour at 40 km, after ~8 hours at 30 km). We note that vertical  
138 motions, including sedimenting ice, also likely contribute to this evolution. Note that the ice plume  
139 persisted the longest (about 20 hours) in the lower stratosphere, near the cold tropopause. Below  
140 the tropopause (~17 km), the cloud drifted in the opposite direction (Fig. 1C) due to prevailing  
141 upper-tropospheric westerlies (Fig. 1B).

142 The young outflow of the eruption was also sampled by MLS, revealing a ~12 km-thick layer  
143 of strongly enhanced water vapour with a top at around 52 km (Fig. 1D) as well as by CALIOP  
144 satellite lidar reporting a strongly depolarizing layer of particles between 35-40 km, just beneath  
145 the moist plume (Fig. 1D). The high depolarization ratio of the plume suggests the presence of  
146 non-spherical particles such as ash and/or ice.



147

148 **Figure 1. Evolution of Hunga volcanic cloud top height (CTH) on the day of eruption (15**  
 149 **January 2022).** (A) (A) East-West sections of maximum CTH color-coded by time from  
 150 stereoscopic retrieval using Himawari-8 and GOES-17 geostationary imagers. (B) ECMWF  
 151 temperature and zonal wind profiles averaged over  $5^\circ \times 5^\circ$  box centred at the volcano location.  
 152 (C) Hovmöller diagram of the maximum CTH (note the inverted time axis). Superimposed lines  
 153 are color-coded by altitude and represent linear trajectories released from the location of volcano  
 154 at different heights indicated in the panel. The circles colour-coded by altitude indicate the  
 155 detections of water vapour and aerosol plumes respectively by MLS and CALIOP. The black-  
 156 rimmed circles indicate the detections of hydrated layers by COSMIC-2 (Fig. S2B, Supplementary  
 157 notes). Note the colour correspondence between the trajectories and downwind detections of the  
 158 plume confirming the CTH retrieval. (D) Latitude-altitude cross section of water vapour from MLS  
 159 (colour map) and depolarization ratio from CALIOP (contours, first contour is 0.05, interval is  
 160 0.05, last contour is 0.25). The time and longitude of MLS and CALIOP plume measurements are  
 161 given in (C).  
 162

163 **Early evolution of volcanic cloud**

164 The explosive eruptive transport together with sedimentation and sublimation of ice produced  
165 a multitude of moist and aerosol-rich layers throughout the depth of the stratosphere. Their  
166 spatiotemporal evolution during the first days after eruption (D+2 to D+4), as observed by MLS  
167 (Fig. 2A, B, C), reveals a wind shear-shaped slant column of moisture extending throughout the  
168 stratospheric layer and spanning from Australia to Africa already on D+2. The strongly hydrated  
169 patches were accompanied by aerosol layers detected by OMPS-LP satellite instrument at all  
170 altitudes between the tropopause (~17 km) and 42.5 km. The presence of aerosols up to 37 km is  
171 confirmed by lidar measurements at La Reunion island downwind of Hunga on D+4 (Fig. 2C and  
172 S5B), which is the highest-level aerosol plume ever observed by ground-based lidars.

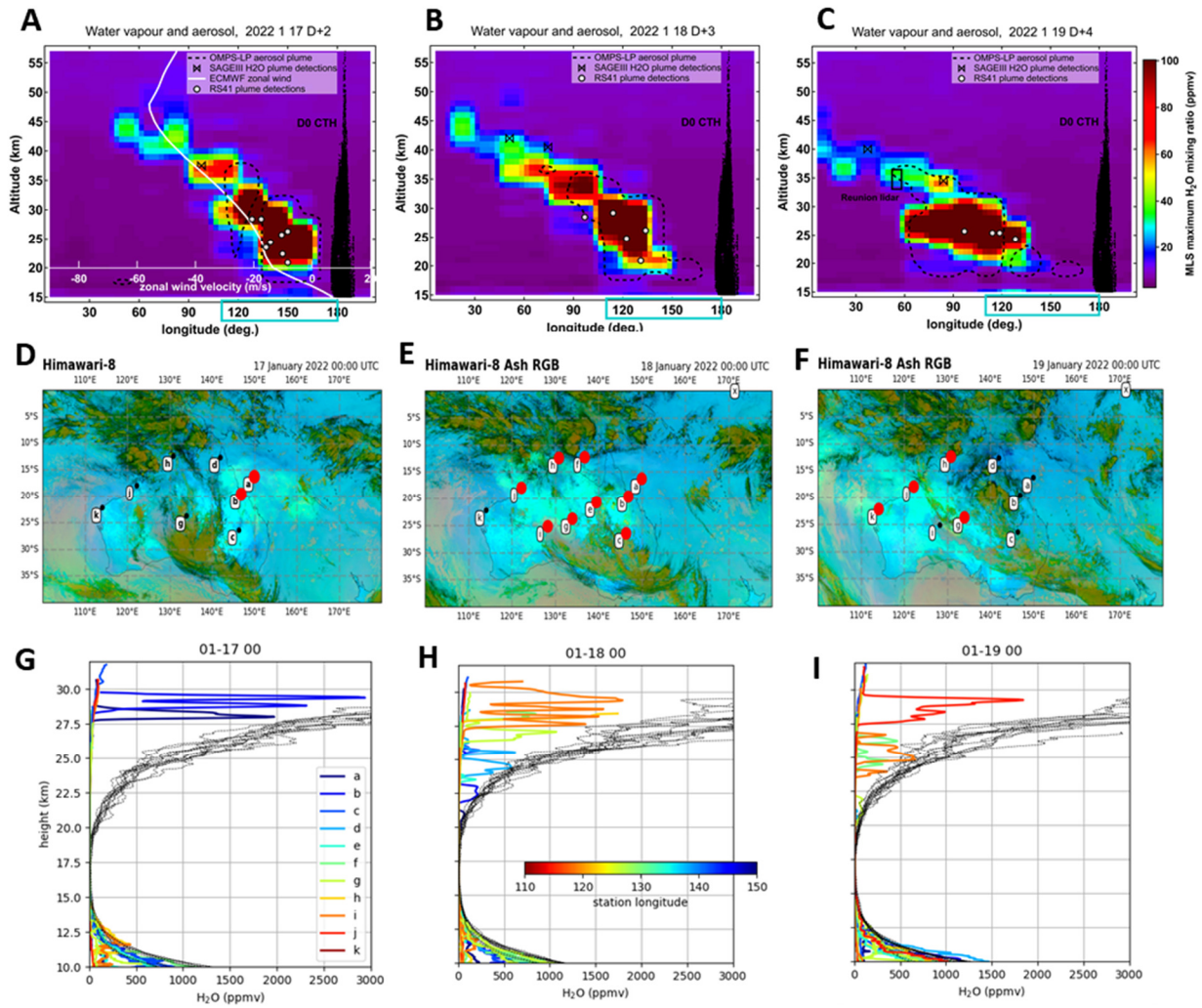
173 The primary volcanic cloud at lower altitudes (<~30 km), traceable by Himawari-8 volcanic  
174 RGB retrieval (Fig. 2D, E, F) was extensively sampled by the Australian upper-air meteorological  
175 network. The radiosondes showed numerous moist layers between 20 - 30 km with peak water  
176 vapour mixing ratios increasing with altitude from around 100 ppmv at 21 km to 2900 ppmv at 28  
177 km following the physical limit of ice saturation at the given level (Fig. 2G, H, I).

178 The presence of large amounts of water in the volcanic plume has probably led to very fast  
179 oxidation of volcanic sulfur dioxide emissions to sulfuric acid (Zhu et al., 2022) - the main  
180 component of stratospheric aerosol droplets. According to CALIOP depolarization measurements,  
181 the aerosol particles were mostly spherical since D+1 and could therefore be characterized as  
182 sulfate aerosol droplets (Legras et al., 2022).

183 The primary aerosol plume at 27-30 km altitude, overpassing La Reunion island on D+6 -  
184 D+7, was marked by an unprecedentedly high optical depth of 0.8 and scattering ratio up to 280  
185 at 532 nm (Baron et al., 2022) (Fig. S5B), which to our knowledge represents the most intense  
186 stratospheric aerosol plume ever observed by ground-based lidars.

187





188

189 **Figure 2. Early evolution of volcanic plume during 17-19 January 2022 (D+2 – D+4).** (A, B,  
 190 C) Longitude-altitude section of MLS maximum water vapour mixing ratio (WVMR) between  
 191 30S – 10S on the respective day. Black points indicate the ice cloud top height on the day of  
 192 eruption (D0). The white curve represents the zonally-averaged ECMWF zonal wind profile. Black  
 193 contours mark the areas where OMPS-LP detected aerosol layers with extinction ratio above 5.  
 194 Diamonds and circles mark the detection of WVMR enhancements above 50 ppmv respectively  
 195 by SAGE III and meteorological Vaisala RS41 radiosoundings. (D, E, F) Locations of the  
 196 Australian radiosounding stations (marked a-k) superimposed on Himawari-8 Ash RGB images  
 197 showing the extent of the primary volcanic plume as aquamarine area. The stations marked red  
 198 represent the detections of WVMR enhancements. (G, H, I) Radiosonde profiles bearing WVMR  
 199 enhancements above 50 ppmv on the respective day (marked by station and color-coded by  
 200 longitude) and corresponding saturation mixing ratio profiles.  
 201

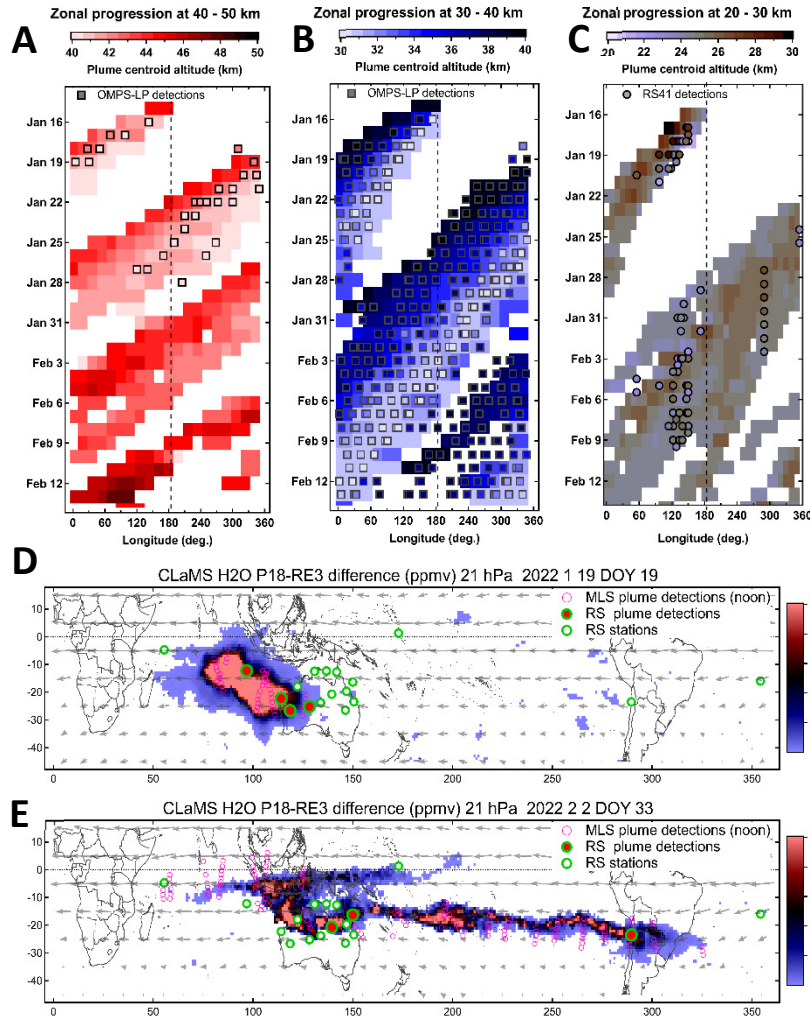
## 202 **Fast circumglobal transport and vertical motion of moisture and aerosols**

203 The extreme altitude reach of the Hunga eruption has led to an unusually fast circumglobal  
204 transport of volcanic material entrained by strong zonal winds in the upper stratosphere, up to 60  
205 m/s at 47 km altitude (Fig. 2A). Consequently, the uppermost plume of moisture circumnavigated  
206 the Earth in only one week and made three full circles in 25 days whilst ascending through the  
207 Brewer-Dobson circulation from ~43 km to ~49 km (Fig. 3A), that is approximately 200 m per  
208 day. The aerosol plume above 40 km has travelled around the globe in 9 days and only made a  
209 single round. The limited lifetime of aerosols at this level could be due to sedimentation and/or  
210 evaporation of sulfate particles in the warm upper stratospheric environment (Kremser et al.,  
211 2016). We note though that the nature of the upper-stratospheric aerosols is unknown due to the  
212 absence of CALIOP depolarization measurements above 40 km.

213 In the middle layer (30 - 40 km), the aerosol and moisture plumes travelled in close tandem  
214 for about a month, circumnavigated the globe in 9 days and entirely covered the Southern tropical  
215 band by 29 January, that is in two weeks (Fig. 3B). Such a fast circumnavigation of the tropical  
216 stratosphere by a volcanic plume is remarkable compared to other major eruptions: the  
217 stratospheric plumes produced by 1982 El Chichon (Robock and Matson, 1983) and 1991 Pinatubo  
218 (Bluth et al., 1992) eruptions had circled the globe in three weeks, although their plumes were  
219 mostly confined to lower altitudes.

220 The zonal progression of the bulk of the plume contained within the 20-30 km layer is found  
221 to be fully consistent between MLS and radiosoundings, both showing a complete  
222 circumnavigation in two weeks (Fig. 3C). During its first circumnavigation, the plume undergoes  
223 significant subsidence, as seen from MLS and radiosounding data (Fig. 3C). We estimate an  
224 average descent rate around 200 m/day with maximum plume top altitudes decreasing from near  
225 30 km during the first overpass over Australia (January 16-19) to ~26 km during the second  
226 overpass (February 1-10). Sellitto et al. (2022) proposed that this vertical motion be driven by  
227 radiative cooling induced by the large water vapor anomaly.

228



229

230 **Figure 3. Circumglobal transport and morphological evolution of hydrated plumes.** (A)  
 231 Evolution of the water vapour mixing ratio (WVMR) peak altitude for the hydrated layers  
 232 (WVMR>10 ppmv) in the upper stratosphere (40 – 50 km) as a function of time and longitude.  
 233 The black squares with altitude-dependent colour meshing indicate the detections of aerosol layers  
 234 with extinction ratio (ER>0.25) by OMPS-LP within the respective altitude range. Note that the  
 235 uppermost plume at around 45 km circumnavigates the globe in only one week. (B) Same as A but  
 236 for the hydrated layers (WVMR>10 ppmv) and aerosol layers (ER>2.5) in the middle stratosphere  
 237 (30 – 40 km). (C) Same as (B) but for the lower tropical stratosphere (20 – 30 km) and with  
 238 detection threshold of 30 ppmv. The black circles with altitude-dependent colour meshing indicate  
 239 the radiosonde detections of WVMR enhancements above 50 ppmv. (D, E) Geographical extent  
 240 of the hydrated plume during its first (D) to second (E) overpass above Australia from CLaMS  
 241 model simulation. The values represent the WVMR difference between the control (pre-eruption  
 242 initialization) and perturbed (post-eruption) simulations exceeding 3 ppmv at 21 hPa level (see  
 243 Methods). The magenta open circles indicate the locations of MLS hydrated layers at 21 hPa with  
 244 WVMR>30 ppmv. The green open circles show the locations of radiosounding stations involved  
 245 in the analysis. The red-filled circles indicate the radiosonde detections of hydrated layers on the

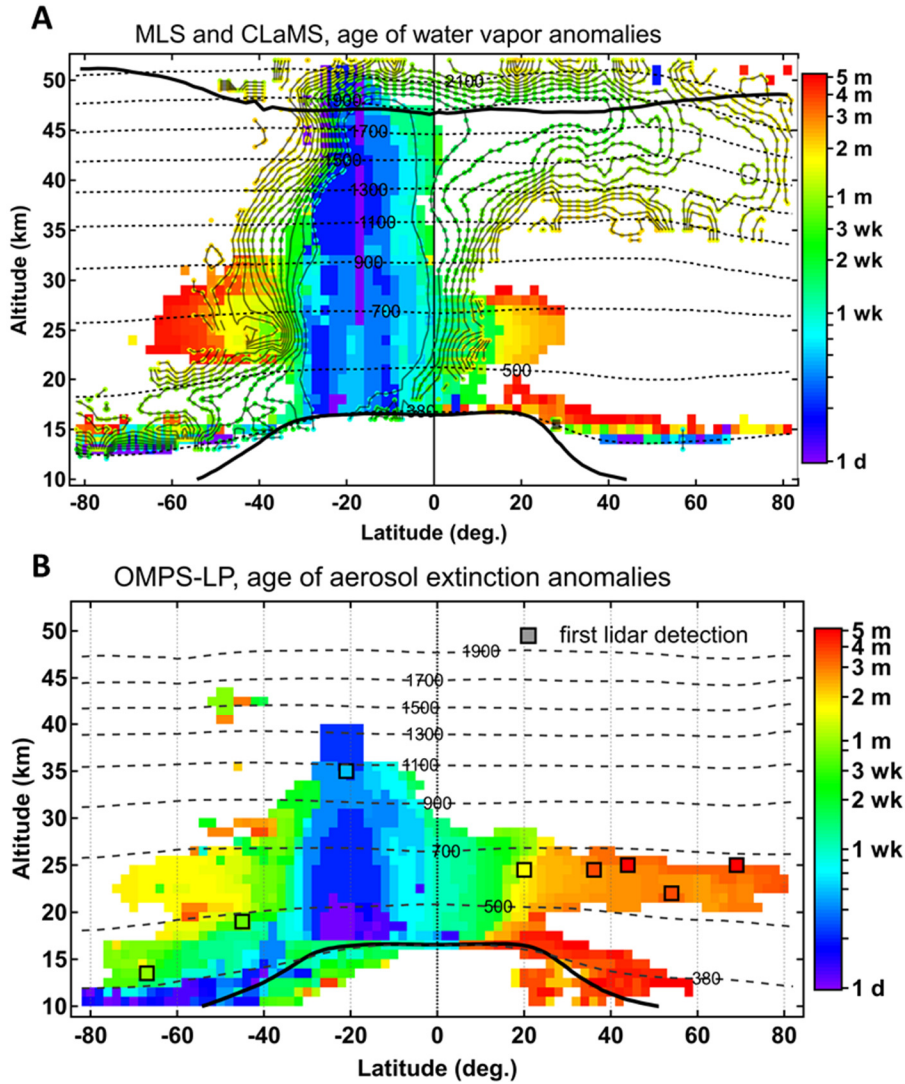
246 respective day. ECMWF wind field at 21 hPa is shown in grey arrows. See Movie S2 for the  
247 complete sequence.

248  
249 Further insight into the morphological evolution of the volcanic moist plumes is provided by  
250 simulations with the CLaMS chemistry-transport model (McKenna et al., 2002) initialised with  
251 MLS water vapour observations (Supplementary notes). The simulation reveals a relatively  
252 compact bulk plume during its first Australian overpass on D+4 (Fig. 3D), whereas during the  
253 second overpass on 2nd February the plume appears as a dragon-shaped structure with a head  
254 emerging around 10° S, where the Easterlies are strongest, and a tail at around 20° S extending  
255 all across the Pacific (Fig. 3E and movie S2). The model simulated plume location, extent and  
256 circumglobal transport is in good agreement with MLS satellite observations (Fig. 3 D-E, pink  
257 circles; for further details see Supplementary notes). The cross-Pacific extent of the bulk plume by  
258 the time of its first circumnavigation is also largely consistent with radiosonde detections of  
259 hydrated layers (Fig. 3E).

260

### 261 **Meridional dispersion of volcanic plumes**

262 After being injected into the southern tropical stratosphere, the volcanic material is  
263 subsequently transported in the meridional plane into Northern and Southern hemispheres by the  
264 stratospheric circulation on a timescale of weeks to months (Fig. 4). The CLaMS simulation  
265 vividly shows the transport towards the North pole along the deep branch of the Brewer-Dobson  
266 circulation (BDC) on a timescale of 1-2 months as well as a fast isentropic transport towards the  
267 South pole in the lowermost stratosphere (Fig. 4A). These pathways are consistent with the known  
268 seasonality of the stratospheric BDC, with the deep branch circulation maximising in hemispheric  
269 winter and the shallow branch circulation maximising in hemispheric summer (e.g., Konopka et  
270 al., 2015). The MLS observations show that within five months of the eruption, the hydrated  
271 plumes have spread in both directions from 65° S to 35° N but mostly within the bulk plume layer  
272 (20 - 30 km). With that, the deep BDC transport pathway is not captured by MLS. These  
273 differences between model and observations regarding meridional transport could be due to the  
274 sensor sensitivity limits in the upper stratosphere and/or due to uncertainties in initial injection  
275 height as represented in the model as well as due to uncertainty in the heating rates, which were  
276 altered due to radiative cooling in the stratosphere induced by excessive moisture (Sellitto et al.,  
277 2022).



279

280 **Figure 4. Global dispersion of water vapour and aerosol plumes during five months since**

281 **Hunga eruption.** (A) Poleward dispersion of hydrated plumes detected by MLS with WVMR

282 climatological anomalies exceeding 3 ppmv. The pixels are colour-coded by the age of hydrated

283 layers since 15 January 2022. The contours (age colour-coding) represent the results of CLaMS

284 model simulation of hydrated plumes transport (WVMR anomalies above 3 ppmv). Thick solid

285 curves mark the tropopause and the stratopause, thin dashed curves indicate isentropic levels. (B)

286 Same as A but for OMPS-LP detections of aerosol layers with extinction ratios exceeding 3. The

287 black rectangles with age-dependent colour meshing indicate the first aerosol layer detections by

288 ground-based lidars (see Fig. S5 and Supplementary notes).

289

290 The meridional dispersion of aerosol plumes (Fig. 4B), as inferred from OMPS-LP  
291 measurements, exhibits prominently the various transport pathways: the fast transport in the  
292 lowermost stratosphere towards the South pole (3-4 weeks), the transport along the BDC between  
293 500 K - 700 K isentropes into both hemispheres (1 - 2 months), which is followed by subsequent  
294 dispersion from the Northern tropics towards the North pole (3 - 4 months). In the lower  
295 stratosphere two distinct pathways emerge, likely involving confinement by the Asian and  
296 American monsoon anticyclones resulting in the gap between the lower and upper branches. The  
297 eventual occurrence of aerosols below the zonally-averaged tropopause level in the Northern and  
298 Southern subtropics suggests sedimentation of large sulfate particles out of the stratosphere.

299 The satellite-derived meridional transport timescale is confirmed by ground-based lidar  
300 detections of aerosol layers (Fig. S5, Supplementary notes) shown as black squares in Fig. 4B. The  
301 fast transport towards the South pole within the lowermost stratosphere is captured by lidars at  
302 Lauder station, New Zealand (45° S) and Dumont d'Urville French Antarctic station (67° S)  
303 respectively 3 and 4 weeks after the eruption. The dispersion of sulfates to the Antarctic region  
304 has thus occurred before the polar vortex formation, and the Dumont d'Urville lidar measurements  
305 report stratospheric aerosol layers at the edge of the vortex in early June (not shown). The  
306 northbound dispersion of aerosols is captured by lidar detections in the northern tropics (Mauna  
307 Loa, Hawaii), subtropics (Tsukuba, Japan), mid-latitudes (OHP, France and Kuhlungsborn,  
308 Germany) as well as high-latitudes (Alomar, Norway). Overall, in about three months since the  
309 eruption, the Hunga sulfates have spread nearly pole-to-pole, although the aerosol layers detected  
310 in the Northern extratropics are less intense, with scattering ratios below 1.8 (Fig. S5).

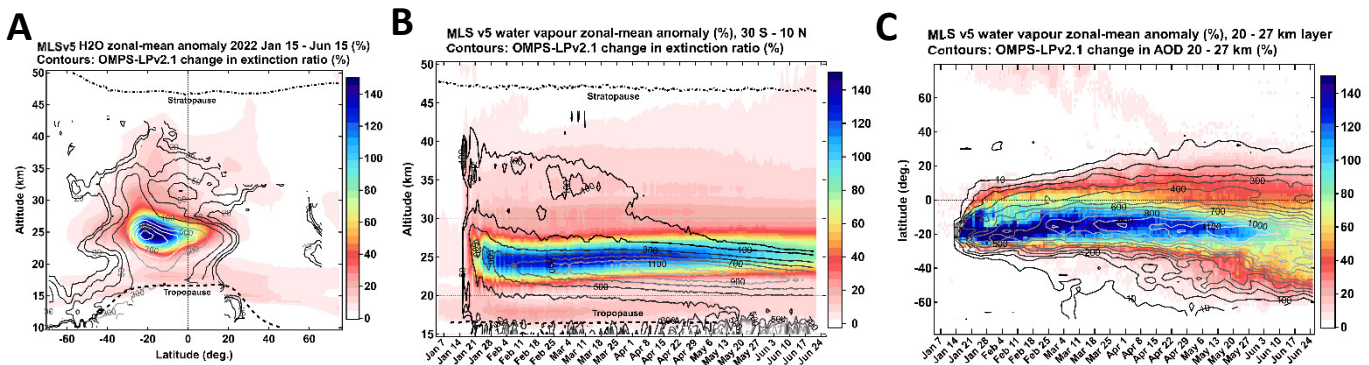
311

### 312 **Global perturbation of stratospheric water and aerosol burden**

313 The extreme explosiveness of the Hunga eruption has led to in-depth perturbations of  
314 stratospheric gaseous and particulate composition. Figure 5A shows the broad-range positive post-  
315 eruption water vapour anomaly (colours) that extends throughout the depth of the tropical  
316 stratosphere - from the tropopause to nearly the stratopause. The region of highest anomalies,  
317 exceeding 100% on a zonal-mean scale and averaged over 5 months after the eruption, is found in  
318 the southern tropics within the 23 - 27 km altitude layer. The anomaly in aerosol extinction  
319 (contours) exceeding 100% extends across most of the tropical lower and middle stratosphere and  
320 reaches 1000% at 24-25 km altitude in the southern tropics. The latitudinal pattern of the aerosol



321 extinction anomaly is well correlated with that of water vapour, except for the downward shift of  
 322 aerosol anomalies. Indeed, while the bulk layer of gaseous water and the upper boundary of the  
 323 positive water anomaly are both gradually rising in the tropical upwelling branch of the Brewer-  
 324 Dobson circulation, the bulk of aerosols is sedimenting with a vertical rate estimated as 0.26 mm/s  
 325 (Fig. 5B, Fig. S7F). The subsidence and meridional dispersion of the bulk aerosol layer is well  
 326 captured by Aeolus ALADIN satellite lidar (Fig. S10). Despite the vertical decoupling of bulk  
 327 water vapour and aerosol layers in the stratosphere, their meridional dispersion reveals a very  
 328 similar pattern with a more efficient transport towards the Southern pole (Fig. 5C).



329  
 330 **Figure 5. Spatiotemporal structure of the stratospheric water vapour and aerosol burden**  
 331 **perturbation.** (A) MLS zonal-mean WVMR anomaly above 5% averaged over five months  
 332 following the eruption with respect to MLS 17-yr climatology (% , color map) and OMPS-LP  
 333 extinction ratio anomaly with respect to pre-eruption conditions (% , contours). (B) Same as A but  
 334 as a function of time for the latitude band 30° S – 10° N. (C) Time-latitude variation of WVMR  
 335 anomaly within 20-27 km altitude layer (color map) and change in OMPS-LP AOD with respect  
 336 to the pre-eruption levels within the same layer (contours).  
 337

338 Figure 6A shows the annual cycle of stratospheric water vapour mass (between 100 hPa - 1  
 339 hPa pressure levels), which is characterised by a minimum in Boreal Spring and a maximum in  
 340 Austral spring with a peak-to-peak amplitude of 60 Tg. The Hunga eruption occurred at the  
 341 midpoint of the decay phase and boosted the stratospheric water burden by  $119 \pm 6$  Tg, which is  
 342 nearly twice the annual amplitude. This figure is fully consistent with the GNSS radio occultation-  
 343 based mass estimate of 100 - 150 Tg. Using an older V4 version of MLS data we obtain the mass  
 344 of water transported across the 100 hPa level of  $137 \pm 7$  Tg, which is consistent with earlier  
 345 estimates of  $139 \pm 8$  Tg by Xu et al. (2022) and  $146 \pm 5$  Tg by Millan et al. (2022). The stratospheric  
 346 water burden perturbation by the Hunga eruption is about a factor of 5 larger than the previous

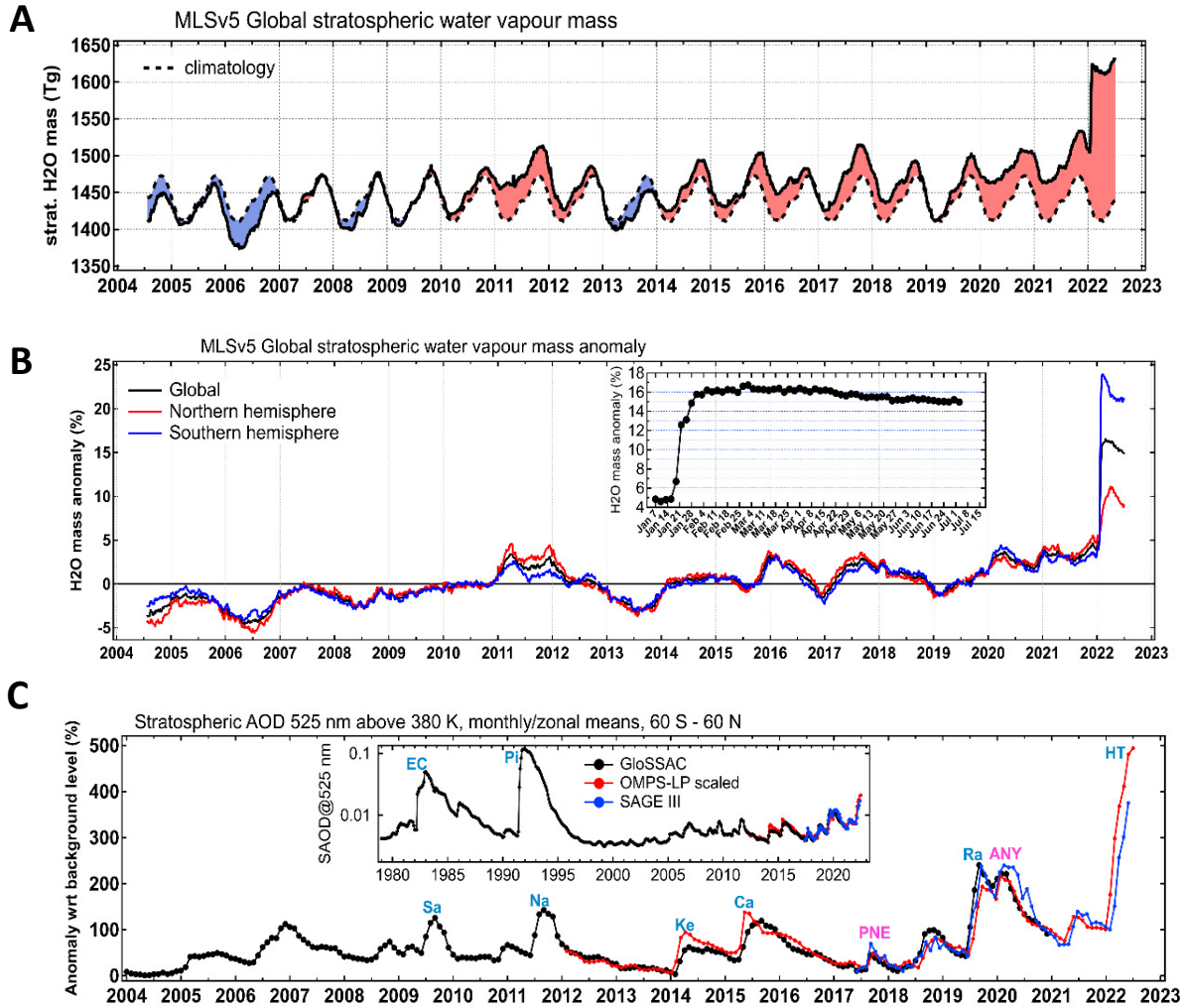
347 record-breaking perturbation of stratospheric water vapour (27 Mt) by the Australian “Black  
348 Summer” wildfires in 2019/2020 (Khaykin et al., 2020).

349 Figure 6B shows that the stratospheric water vapour mass anomaly has reached ~24% in  
350 the Southern and ~11% in the Northern hemisphere, whereas the global anomaly has reached ~16%  
351 after the eruption. Note that over the 17-yr time span of MLS data, the global and hemispheric  
352 anomalies do not exceed 5%, which renders the Hunga-induced perturbation of stratospheric water  
353 load unique in the record. Indeed, the Stratospheric Water and OzOne Satellite Homogenized data  
354 set (SWOOSH) (Davis et al., 2016) including satellite measurements of stratospheric water vapour  
355 since 1985, clearly shows that the perturbation is unprecedented in the satellite record of  
356 stratospheric water vapour.

357 The underwater blast associated with Hunga eruption and subsequent ice-vapour phase  
358 transition in the stratosphere led to a substantial increase in heavy water isotopologues as inferred  
359 from ACE-FTS data (Fig. S9, Supplementary notes). The extreme excursion of water isotopic ratio  
360 towards the Standard Mean Ocean Water (SMOW) levels strongly suggests seawater as the main  
361 source of injected moisture.

362 In order to place the stratospheric aerosol load perturbation by Hunga into historical  
363 perspective, we combined the GloSSAC merged satellite dataset (Kovilakam et al., 2020) spanning  
364 1979-2020 with the recent aerosol extinction measurements by OMPS-LP and SAGE III satellite  
365 sensors. Figure 6C provides evidence that the Hunga eruption led to a 4-5-fold increase in the  
366 stratospheric aerosol optical depth (SAOD), exceeding by far any volcanic or wildfire event in the  
367 last three decades. With that, the absolute magnitude of SAOD perturbation (embedded panel in  
368 Fig. 6C) by Hunga is at least a factor of 6 smaller than to the previous major eruption of Pinatubo  
369 in 1991 and factor of 3 smaller than that of El Chichon in 1982.





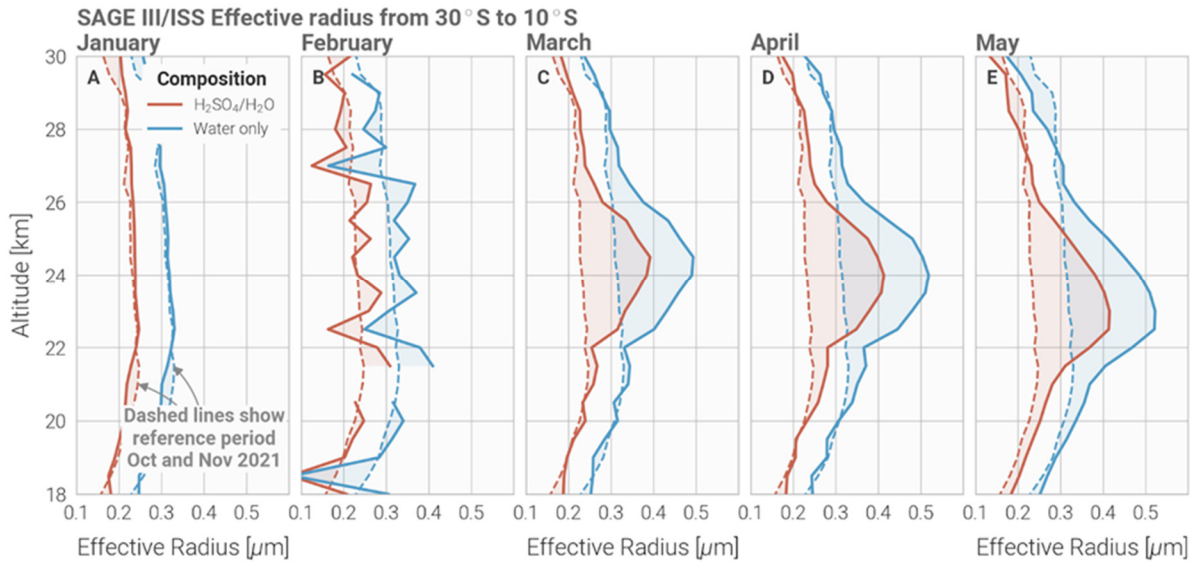
370

371 **Figure 6. Global perturbation of stratospheric water vapour and aerosol burden.** (A)  
 372 Evolution of the global MLS stratospheric water vapour mass (3-day averages) between 100 hPa  
 373 – 1 hPa pressure levels (solid black curve) and climatological (2004-2021 period) annual cycle  
 374 (dashed curve), the positive and negative anomalies are shown respectively as red and blue  
 375 shading. (B) Deseasonalized stratospheric water vapour mass anomaly (per cent 3-day averages)  
 376 for both hemispheres and the whole globe from MLS. The embedded panel shows the evolution  
 377 of global anomaly in 2022. (C) Stratospheric aerosol optical depth (SAOD) anomalies for the 60°  
 378 S – 60° N latitude band (monthly averages) from GloSSAC merged satellite record extended using  
 379 OMPS-LP measurements at 675 nm scaled to 525 nm wavelength using GloSSAC data and SAGE  
 380 III/ISS measurements at 521 nm converted to 525 nm using SAGEIII-derived Angstrom exponent.  
 381 The SAOD anomalies are computed with respect to the background level estimated as GloSSAC  
 382 SAOD average over volcanically-quiescent 1995-2003 period. The embedded panel shows the full  
 383 time span of SAOD series. The cyan and pink letters indicate the most significant volcanic  
 384 eruptions and wildfire events respectively (EC – El Chichon, Pi – Pinatubo, Sa – Sarychev, Na –  
 385 Nabro, Ke – Kelud, Ca – Calbuco, PNE – Pacific Northwest wildfire event, Ra – Raikoke, ANY  
 386 – Australian New Year wildfire event, HT – Hunga Tonga).

387 Of particular interest is the post-eruption evolution of stratospheric aerosol size. Figure 7  
388 shows the effective radius retrieved from SAGE III for the months following the Hunga eruption  
389 using different assumptions on the aerosol composition. Background conditions, shown in dashed  
390 lines, typically have an effective radius of approximately 230 nm. After the eruption, particle size  
391 increases from the background values to over 400-500 nm, depending on composition; larger than  
392 at any other point in the SAGE III/ISS record. This growth is contained primarily between 22 and  
393 26 km, which contains the bulk of the enhanced aerosol. By mid-March the particles have reached  
394 their maximum size and this layer begins settling. A more detailed analysis of retrieved size  
395 parameters suggests a complex interplay between the sedimentation, condensation and coagulation  
396 processes (Supplementary notes, Fig. S8).

397 The sedimentation rate of aerosol was estimated from OMPS-LP tomographic retrieval of  
398 extinction profiles by tracking the peak altitude of the plume (Supplementary notes and Fig. S7).  
399 A linear fit to the peak beginning March 10th suggests a settling rate of 0.26 mm/s, which is in  
400 agreement with CALIOP-derived estimates by Legras et al. (2022). Taking into account the  
401 monthly averaged ERA5 vertical wind speed we obtain the fall speed, based on which the particle  
402 size can be estimated using the method proposed by Kasten (1968). Depending on the assumption  
403 on the particles' relative fraction of  $\text{H}_2\text{O}/\text{H}_2\text{SO}_4$ , we obtain a radius of 350 to 540 nm in April-  
404 May, which is fully consistent with the SAGE III-derived particle sizes, providing confidence in  
405 these estimates.

406



407  
 408 **Figure 7. Estimation of particle size from SAGE III/ISS multiwavelength data.** Panels A-E  
 409 show the monthly averaged retrieved effective radius from SAGE III/ISS assuming background  
 410 (red) and pure-water (blue) aerosol composition. Dashed lines indicate the average effective radius  
 411 pre-eruption, computed from October and November 2021 profiles.

412

### 413 Discussion

414 The extreme explosiveness of the Hunga eruption and the submarine location of the volcano  
 415 add up to the unprecedented character, magnitude and the propagation timescale of the global  
 416 stratospheric perturbation. The eruption provided a unique natural testbed, lending itself to studies  
 417 of climate sensitivity to strong change in both stratospheric gaseous and particulate composition.  
 418 In particular the effect on stratospheric water vapour is tremendous and expected repercussions  
 419 range from persistent changes in atmospheric radiative balance (Santer et al., 2014; 2015) to  
 420 amplification of the polar ozone depletion through wider occurrence of polar stratospheric clouds  
 421 (Zhu et al., 2022).

422 The high persistence of the perturbation, related to the high injection altitude and extreme  
 423 amount of stratospheric moisture together with the significant amount of sulfates generated by the  
 424 Hunga eruption, will likely cause particularly long-lasting perturbations to atmospheric radiation  
 425 and stratospheric chemistry. In six months since the eruption, the global anomaly of the  
 426 stratospheric water burden did not decay more than 1-2%, and such persistence is expected

427 considering the absence of water vapour sinks in the middle stratosphere, that is where the bulk  
428 layer of the Hunga moisture is contained.

429 While the longer-term aftermath of the Hunga effects is yet to be known, the available data  
430 provide enough evidence to rank this eruption among the most remarkable climatic events in the  
431 modern observational era and strongest in the last three decades. As remote sensing techniques  
432 and satellite coverage of the stratosphere have been substantially improved in the XXI century, the  
433 wealth of observational data on the Hunga event together with various modelling approaches  
434 should provide a major advance in understanding the impacts of stratospheric composition change  
435 on global climate.

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448 **Author contributions:** SK conceived the study and performed analysis of MLS, OMPS-LP,  
449 CALIOP, SAGEIII, GLoSSAC and CLaMS data; AP performed analysis of radiosoundings and  
450 COSMIC-2 data, FP and JUG performed CLaMS simulation and analysis of MLS data, KK and  
451 KB performed stereoscopic CTH retrieval; FT and SB computed stratospheric water vapour  
452 masses; BLe provided Ash RGB data; LR computed particle radius from SAGEIII and OMPS-LP  
453 data; TS, JB, OU, IM, TN, RW, GB, MG, AB, VD, GP, JJ, RQ, BLi, AH provided processed lidar  
454 data; AF performed analysis of Aeolus data; BC performed analysis of ACE-FTS data; Ble, AP,  
455 PS, SB, SGB, FR, AB were involved in discussions of the results and their interpretation. All  
456 authors contributed to the final manuscript. The paper was written by SK, AP, FP, SB, LR, SGB,  
457 JUG.

458 **Competing interests:** Authors declare that they have no competing interests.

459 **Data and code availability:** MLS water vapour data are available at  
460 [https://acdisc.gesdisc.eosdis.nasa.gov/data/Aura\\_MLS\\_Level2/ML2T.005/2022/](https://acdisc.gesdisc.eosdis.nasa.gov/data/Aura_MLS_Level2/ML2T.005/2022/). CALIOP data  
461 v3.41 are available at: [https://doi.org/10.5067/CALIOP/CALIPSO/CAL\\_LID\\_L1-](https://doi.org/10.5067/CALIOP/CALIPSO/CAL_LID_L1-VALSTAGE1-V3-41)  
462 [VALSTAGE1-V3-41](https://snpp-omps.gesdisc.eosdis.nasa.gov/data/SNPP_OMPS_Level2/OMPS_NPP_LP_L2_AER_DAILY.2/2022/). OMPS V2.0 data is available at [https://snpp-](https://snpp-omps.gesdisc.eosdis.nasa.gov/data/SNPP_OMPS_Level2/OMPS_NPP_LP_L2_AER_DAILY.2/2022/)  
463 [omps.gesdisc.eosdis.nasa.gov/data/SNPP\\_OMPS\\_Level2/OMPS\\_NPP\\_LP\\_L2\\_AER\\_DAILY.2/](https://snpp-omps.gesdisc.eosdis.nasa.gov/data/SNPP_OMPS_Level2/OMPS_NPP_LP_L2_AER_DAILY.2/2022/)  
464 [2022/](https://snpp-omps.gesdisc.eosdis.nasa.gov/data/SNPP_OMPS_Level2/OMPS_NPP_LP_L2_AER_DAILY.2/2022/); OMPS-LP V2.1 data is available at  
465 [https://avdc.gsfc.nasa.gov/pub/data/satellite/Suomi\\_NPP/L2/LP-L2-AER-45km/LP-L2-AER-](https://avdc.gsfc.nasa.gov/pub/data/satellite/Suomi_NPP/L2/LP-L2-AER-45km/LP-L2-AER-)

466 DAILY/2022/; OMPS-LP tomographic retrieval data are available at ftp://odin-osiris.usask.ca/  
467 with login/password osirislevel2user/hugin; SAGE III data at  
468 [https://doi.org/10.5067/ISS/SAGEIII/SOLAR\\_BINARY\\_L2-V5.2](https://doi.org/10.5067/ISS/SAGEIII/SOLAR_BINARY_L2-V5.2); Aeolus data are available at  
469 [https://aeolus-ds.eo.esa.int/oads/access/collection/Level\\_2A\\_aerosol\\_cloud\\_optical\\_products/](https://aeolus-ds.eo.esa.int/oads/access/collection/Level_2A_aerosol_cloud_optical_products/);  
470 ERA5 data are available at <https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era5>.  
471 The scripts and notebooks used in this study as well as intermediate datasets will be available  
472 from Zenodo. In the mean time, all the materials used in the study can be obtained by contacting  
473 the corresponding author.

## 474 **Methods**

### 475 *Stereoscopic cloud top height (CTH) retrieval*

476 Two primary steps were used in the derivation of cloud top height for the Hunga Tonga-  
477 Hunga Ha'apai eruption cloud based on GOES-17 and Himawari-8 geostationary satellite  
478 observations: 1) spatially matching simultaneous observations from the two satellites, and 2) using  
479 the stereoscopy principle to construct a 3D profile of the cloud. Because the two satellites have  
480 sufficiently different viewing angles, then it can be possible to derive a cloud top height with  
481 accuracy equal to or better than the spatial resolution of the imagery being used. Level 1B infrared  
482 (IR) brightness temperature (BT) data in the 10.3  $\mu\text{m}$  is collected at 2 km/pixel nadir resolution  
483 every 10 minutes and nearly simultaneously from GOES-17 and Himawari-8 because the imagers  
484 on these satellites, the Advanced Baseline Imager (ABI) and Advanced Himawari Imager (AHI)  
485 respectively, are nearly identical and have the same scan initiation times and scan rate. Although  
486 IR imagery is of lower resolution than the visible, it has its own advantages as it is free of shadows,  
487 is nearly isotropic, and is available at nighttime. Pixel geolocation in Level 1B data is obtained by  
488 intersecting the instant view axis of the imager instrument with the Earth reference ellipsoid, and  
489 thus the nominal image registration is accomplished assuming a zero elevation of observed scenes.  
490 Once these Level 1B data are reprojected from the satellite's pixel/line space to a geographical  
491 projection, any elevated scene exhibits a parallax displacement, which is different for images  
492 recorded at different viewing angles. With simple geometric transformations, the two parallax  
493 displacements from the two satellites can be directly related to the sought height.

494 An algorithm developed at NASA Langley Research Center uses image subsets (chips)  
495 ranging from 8x8 to 20x20 pixel sizes to obtain a cross correlation between chips from the two  
496 image sources. Trying different relative displacements between the chips consecutively yields the  
497 highest correlation at the position of optimal displacement, which corresponds to the actual height  
498 for that image subset. Analyses indicate that we were able to achieve a subpixel accuracy when

499 calculating the position of the highest correlation. This translates to a typical accuracy of the  
500 derived height on the order of 0.2-0.4 km. When the analyzed image chips have little texture, the  
501 correlation matching may fail for smaller chip sizes. In that case, a larger chip can be used to obtain  
502 a reliable peak in the correlation profile, but that lowers the effective resolution of the resulting  
503 map of retrieved heights. More than 90% of image chips, however, were reliably matched using  
504 the 8x8 chip size, which helps to resolve smaller features and details within the eruption cloud,  
505 like the small peaks of cloud extending above 50 km altitude. Overall, we estimate the spatial  
506 resolution of the cloud top height retrieval product to be ~4-6 km/pixel. This algorithm was applied  
507 to satellite data from 0400 to 2350 UTC on 15 January 2022 to quantify heights reached by the  
508 eruption cloud and document its temporal evolution.

### 509 *COSMIC-2 water vapour retrieval*

510 Constellation Observing System for Meteorology Ionosphere and Climate (FORMOSAT-  
511 7/COSMIC-2) (Schreiner et al., 2020) is a recently launched equatorial constellation of six  
512 satellites carrying advanced GNSS (Global Navigation Satellite System) radio occultation (RO)  
513 receivers, providing high vertical resolution profiles of bending angles and refractivity, which  
514 contain information on temperature and water vapor. A few RO soundings occurred inside the  
515 Hunga plume on January 15 th, depicting extremely unusual large refractivity anomalies. While  
516 refractivity  $N$  in the neutral atmosphere depends on temperature  $T$ , pressure  $P$ , water vapor partial  
517 pressure  $e$  and liquid water  $W$  (Kurskinky et al, 1997, equations 7 and 11):

$$518 \quad N = 77.6 P/T + 3.73 \cdot 10^5 e/T^2 + 1.4 W,$$

519 the magnitude of the anomalies of the profiles in the early plume would result in temperature  
520 anomalies with unphysically low values near the plume. On the contrary volcanic plume studies  
521 suggest that the temperature within the plume relaxes to that of the background atmosphere within  
522 a few tenths of minutes, with differences (e.g., associated with waves) below 10 K amplitude in  
523 the stratosphere. Hence, we assume that the temperature is at environmental values, as given by  
524 the high resolution operational analysis of the European Center for Medium Range Weather  
525 Forecast (ECMWF), and attribute the refractivity anomaly signal solely to water vapor, in  
526 agreement with expectations regarding the adjustment of a volcanic plume (Woods, 1988, Glaze  
527 et al., 1997), which can be retrieved from (Kurskinky et al, 1997, equation 11):

$$528 \quad e = (N T^2 - b_1 P T)/(b_2)$$

529 ***MLS***

530 The MLS (Microwave Limb Sounder) (Waters et al., 2006) instrument on the NASA Aura  
531 satellite has been measuring the thermal microwave emission from Earth's atmospheric limb since  
532 July 2004. With ~15 orbits per day, MLS provides day and night near-global (82° S–82° N)  
533 measurement of vertical profiles of various atmospheric gaseous compounds, geopotential height  
534 and temperature of the atmosphere. The measurements yield around to 3500 profiles per day for  
535 each species with a vertical resolution of ~3–5 km.

536 For this study we use version 5.01 MLS water vapour product. The data are accompanied by  
537 indicators about data quality and the status of the retrieval convergence. As stated by Millan et al.  
538 (2022), most of the early MLS measurements of the Hunga hydrated plume did not pass the MLS  
539 quality screening. Therefore, here we use MLS water vapour data without accounting for the  
540 quality flag as in Millan et al. (2022).

541 The stratospheric mass load of H<sub>2</sub>O was derived from MLS volume mixing ratio  
542 measurements of water vapour in log pressure space, molecular mass of the compound and the air  
543 number density derived from MLS temperature profile on pressure levels between 100 hPa - 1 hPa  
544 levels. The error bars on the mass of injection are estimated by combining accuracies on the  
545 measurements and the mean standard deviations over 20-day periods before and after the sharp  
546 increase. See Supplementary notes for further detail on the mass estimation method.

547

548 ***CALIOP***

549 The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) is a two-wavelength  
550 polarization lidar on board the CALIPSO mission that performs global profiling of aerosols and  
551 clouds in the troposphere and lower stratosphere (Winker et al., 2010). We use the total attenuated  
552 532 nm backscatter level 1 product V3.40. The depolarization ratio is computed as the ratio  
553 between the perpendicular and parallel components of the attenuated backscatter.

554

555 ***OMPS-LP***

556 The Ozone Mapping and Profiler Suite Limb Profiler (OMPS-LP) on the Suomi National  
557 Polar-orbiting Partnership (Suomi-NPP) satellite, which has been in operation since April 2012,  
558 measures vertical images of limb scattered sunlight in the 290-1000 nm spectral range (Jaross et  
559 al., 2014). The sensor employs three vertical slits separated horizontally to provide near-global

560 coverage in 3–4 days and more than 7000 profiles a day. Here we use OMPS-LP V2.0 aerosol  
561 extinction data (Taha et al., 2021) at 675 nm for analysis of long-term stratospheric AOD evolution  
562 (Fig. 6) and a special time period-limited V2.1 data version extended to 45 km altitude for the rest  
563 of the analysis. Extinction ratio is computed as the ratio between aerosol and molecular extinction.  
564 For estimating the sedimentation rate of aerosol particles, we use OMPS-LP data retrieved using  
565 a tomographic algorithm (Zawada et al., 2018), which provides extinction profiles at 755 nm with  
566 1- 2 km resolution throughout the stratosphere.

567

### 568 *Meteorological radiosoundings*

569 We use the data of meteorological radiosoundings conducted with high-accuracy Väisälä  
570 RS41 sondes in the Southern tropics (Australia, Saint Helena island, Seychelles, Chile and  
571 Argentina). Under normal circumstances, stratospheric humidity is particularly difficult to  
572 measure due to low ambient relative humidity and large outgassing from the balloon (~100 ppmv  
573 at 30 km) overwhelming the small stratospheric water signal (e.g. Vömel et al., 2007). However,  
574 this contamination is outweighed by the ultra-moist plume HT plume which clearly stands out  
575 from background variability and exceeds uncertainties of Vaisala RS41 during its first round-the-  
576 globe tour. The humid plume (RH>20%) within the relatively warm upper stratosphere (T>220 K)  
577 constitutes a favorable environment for RS41 humidity measurements (Survo et al., 2015). While  
578 only RS41 data are included in this survey, other lower resolution sondes detected the plume during  
579 its first overpass (Vaisala RS92) whereas others did not exhibit significant enhancements (M10,  
580 iMET).

581 Later on during the plume dispersion, the raw water vapor signal is diluted and becomes  
582 difficult to isolate from the effect of altitude-dependent outgassing. It is nevertheless possible to  
583 track the plume as an anomaly from a typical contamination profile defined as the 90% quantile  
584 profile over one month for each station. This simple approach is sufficient for the second overpass  
585 over continental stations but fails over tropical islands where outgassing exhibits significant  
586 variability related to low level moisture and cloudiness profile.

587

### 588 *Himawari-8 Ash RGB*

589 The early stage evolution of the plume is tracked with a composite RGB product that benefits  
590 from the sensitivity of the Himawari-8 8.5  $\mu\text{m}$  band to SO<sub>2</sub> and sulphate aerosols. The product is



591 based on the EUMETSAT Ash RGB recipe and uses the brightness temperatures (BT in K) of the  
592 three channels: 8.5, 10.4 and 12.3  $\mu\text{m}$ . The recipe for the three colour indexes ranging from 0 to 1  
593 is  $R = (\text{BT}(12.3) - \text{BT}(10.4) + 4)/6$ ,  $G = (\text{BT}(10.4) - \text{BT}(0.85) + 4)/9$ ,  $B = (\text{BT}(10.4) - 243)/60$ . This  
594 product qualitatively distinguishes thick ash plumes or ice clouds (brown), thin ice clouds (dark  
595 blue) and sulphur-containing plumes (neon-green). Mixed ash/sulphur-containing volcanic species  
596 would appear in reddish and yellow shades. We stress that this satellite product cannot distinguish  
597 between  $\text{SO}_2$  and sulphate aerosols, which have overlapping spectral signatures in this spectral  
598 range (Sellitto et al., 2017) and both appear as neon-green.

599

### 600 *CLaMS chemistry-transport model simulation*

601 The evolution of the Hunga Tonga water vapour plume through the stratosphere has been  
602 simulated with the Chemical Lagrangian Model of the Stratosphere, CLaMS (e.g., McKenna et al.,  
603 2002). CLaMS is a 3d Lagrangian chemistry transport model with transport and chemistry offline  
604 driven by wind and temperature data from meteorological (re)analysis or climate models.  
605 Lagrangian model transport is based on the computation of forward trajectories with an additional  
606 parameterization of small-scale turbulent mixing processes, depending on the shear in the large-  
607 scale flow. The calculation of stratospheric water vapour in CLaMS is based on a freeze-drying  
608 parameterization depending on local saturation mixing ratios along the air parcel trajectories and  
609 a mean sedimentation velocity for ice, and additional chemistry for representing methane oxidation  
610 (e.g. Poshyvailo et al., 2018). This model representation of relevant de- and re-hydration processes  
611 together with CLaMS' Lagrangian transport scheme has been shown advantageous for reliably  
612 simulating the stratospheric water vapour distribution (e.g. Ploeger et al., 2013) and its trend  
613 (Konopka et al., 2022). Also the transport of volcanic plumes has recently been simulated  
614 realistically with CLaMS (Kloss et al., 2021).

615 For this study we used the operational analysis from the European Centre of Medium-range  
616 Weather Forecasts (ECMWF) for driving model simulations. Model transport in the stratosphere  
617 is formulated using a diabatic coordinate in the vertical (potential temperature) and the required  
618 diabatic heating rates have been calculated via a Morcrette scheme assuming clear-sky conditions  
619 (e.g. Konopka et al., 2007). We carried out a control simulation for unperturbed conditions, with  
620 stratospheric water vapour initialised with mixing ratios observed by MLS just before the Tonga  
621 eruption on 13 January 2022, and a perturbed simulation with water vapour initialised just after

622 the eruption on 18 January 2022. For preparing 3D water vapour initialisation fields, MLS  
623 measurements (data version 5) have been mapped onto the closest synoptic time (12 UTC) using  
624 forward/backward trajectories, and have subsequently been binned to a regular 1x3 degree latitude-  
625 longitude grid on the respective MLS pressure levels (see above). Data gaps in these regularly  
626 gridded MLS water vapour distributions related to the coarse satellite sampling have been filled  
627 by interpolation from values around, before using these distributions for initialising the CLaMS  
628 irregularly spaced Lagrangian model grid on 13 and 18 January 2022 via interpolation.

629

### 630 *Ground-based lidars*

631 We use aerosol backscatter measurements at 532 nm provided by ground-based lidars at  
632 various locations to characterize the time scale of the meridional dispersion of Hunga aerosol  
633 plumes. The aerosol plumes are detected as local maxima in scattering ratio exceeding 1.2. The  
634 lidar stations involved in this study (sorted by latitude) are Dumont d’Urville (67° S), Lauder (45°  
635 S), Reunion (21° S), Mauna Loa (20° N), Tsukuba (36° N), Haute Provence (44° N), Kuhlungsborn  
636 (54° N) and Alomar (69° N). The description of the measurement stations and lidar instruments is  
637 provided in Supplementary notes.

638

### 639 *SAGE III*

640 The Stratospheric Aerosol and Gas Experiment (SAGE) III/ISS provides stratospheric aerosol  
641 extinction coefficient profiles using solar occultation observations from the International Space  
642 Station (ISS) (Cisewski et al., 2014). These measurements, available since February 2017, are  
643 provided for nine wavelength bands from 385 to 1550 nm and have a vertical resolution of  
644 approximately 0.7 km. The SAGE III/ISS instrument and the data products have characteristics  
645 nearly identical to those from the SAGE III Meteor mission. We use version V5.2 of SAGE III  
646 solar occultation species data. Particle size is retrieved from SAGE III/ISS by fitting the extinction  
647 spectrum from 384 to 1540 nm using a unimodal lognormal particle size distribution. Typically,  
648 particles in the stratosphere are composed primarily of sulfuric acid and water with a 75/25 mix of  
649 H<sub>2</sub>SO<sub>4</sub>/H<sub>2</sub>O. This assumption impacts the particle size retrieval through the index of refraction,  
650 which for background conditions is typically between 1.40 to 1.44, depending on wavelength. If  
651 particles are more hydrated this may reduce the index of refraction. To estimate the upper bound  
652 of the error due to the assumed index of refraction the retrieval is also performed assuming droplets

653 of pure-water, which leads to retrieved effective radii consistently 100 nm larger than when  
654 particles have a H<sub>2</sub>SO<sub>4</sub>/H<sub>2</sub>O mix.

#### 655 ***GloSSAC merged satellite aerosol climatology***

656 The Global Space-based Stratospheric Aerosol Climatology (GloSSAC) is a 38-year  
657 climatology of stratospheric aerosol extinction coefficient measurements by various satellite  
658 instruments such as SAGE, OSIRIS, CALIOP (Kovilakam et al., 2020). Data from other space  
659 instruments and from ground-based, aircraft and balloon-borne instruments are used to fill in key  
660 gaps in the data set. Here we use GloSSAC V2.1 data on aerosol extinction at 525 nm.

#### 661 ***ALADIN/Aeolus***

662 The European Space Agency's Aeolus satellite carries a Doppler wind lidar called ALADIN  
663 (Atmospheric Laser Doppler INstrument), which operates at 355 nm wavelength and which can  
664 separate the molecular (Rayleigh) and particular (Mie) backscattered photons (high spectral  
665 resolution lidar, HSRL). The lidar observes the atmosphere at 35° from nadir and perpendicular to  
666 the satellite track, its orbit is inclined at 96.97°, and the instrument overpasses the equator at 6h  
667 and 18h of local solar time (LST). We use its L2A Aerosol/Cloud optical product (baseline 12  
668 and above) retrieved with the help of Standard Correct Algorithm (Flament et al., 2021) and  
669 available at 87 km horizontal resolution.

#### 670 ***ACE-FTS***

671 The ACE-FTS (Atmospheric Chemistry Experiment Fourier Transform Spectrometer)  
672 (Boone et al., 2020) is the primary instrument aboard SCISAT. It has been observing about 30  
673 solar occultations per day since 2004, recording spectra between 750 cm<sup>-1</sup> to 4400 cm<sup>-1</sup> at a  
674 spectral resolution of 0.02 cm<sup>-1</sup> and an altitude resolution of 1-2 km. Volumetric mixing ratio  
675 profiles of more than 30 trace gases can be inferred from these spectra, including those of H<sub>2</sub>O and  
676 HDO. In this study we use the Version 4.1/4.2 Level 2 VMR retrievals of H<sub>2</sub>O and HDO. Vapor-  
677 phase deltaD is derived from these quantities at altitude levels between 12 and 40 km. DeltaD is a  
678 measure of the HDO/H<sub>2</sub>O ratio in a sample relative to ratio found in Standard Mean Ocean Water  
679 (SMOW). Flags from both species are used to assess retrieval quality and determine at which  
680 altitude ranges retrievals were actually performed.

681

682 **References**

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