1 2 3	Stratospheric temperature and ozone impacts of the Hunga Tonga-Hunga Ha'apai water vapor injection
4 5 7 8 9	Eric L. Fleming <sup>1,2</sup> , Paul A. Newman <sup>1</sup> , Qing Liang <sup>1</sup> , and Luke D. Oman <sup>1</sup> <sup>1</sup> NASA Goddard Space Flight Center, Greenbelt, MD, USA <sup>2</sup> Science Systems and Applications, Inc., Lanham, MD, USA
10 11 12	Corresponding author: Eric Fleming (eric.l.fleming@nasa.gov)
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15	Key Points:
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17	• The water vapor injection from the Hunga Tonga eruption impacts stratospheric temperature
18	and ozone.
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20	• Largest additional ozone depletion is estimated to occur in Antarctic spring 2023. The ozone
21	response may not be detectable in observations.
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23	• The impacts are projected to gradually diminish after 2024 as the excess water vapor is
24	removed from the stratosphere.
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#### 35 Abstract.

The January 2022 eruption of the Hunga Tonga-Hunga Ha'apai underwater volcano injected a 36 37 large amount of water vapor into the mid-stratosphere. This study uses model simulations to investigate the resulting stratospheric impacts out to 2031. Maximum radiatively-driven model 38 temperature changes occur in the Southern hemisphere (SH) subtropics in April-May 2022, with 39 warming of ~1K in the lower stratosphere and cooling of 3K in the mid-stratosphere. The 40 radiative cooling combined with adiabatic cooling driven by the quasi-biennial oscillation 41 42 meridional circulation explains the near-record cold anomaly observed in the SH subtropical mid-stratosphere. Projected ozone responses maximize in 2023-2024 as the water vapor plume is 43 44 transported globally throughout the stratosphere and mesosphere. The excess H<sub>2</sub>O increases the OH radical, causing a negative global ozone response (2-10%) in the upper stratosphere and 45 46 mesosphere due to increased odd hydrogen-ozone loss, and a small positive ozone response (0.5-47 1%) in the mid-stratosphere due to interference of the NOx catalytic loss cycle by the additional 48 OH. In the lower stratosphere, the excess  $H_2O$  is projected to increase polar stratospheric clouds 49 and springtime halogen-ozone loss, enhancing the Antarctic ozone hole by 25-30 DU in 2023. 50 Arctic impact is small, with maximum additional ozone loss of 4-5 DU projected in spring 2024. 51 These responses diminish after 2024 to be quite small by 2031, as the excess  $H_2O$  is removed 52 from the stratosphere with a 2.5-year e-folding time. Given the year-to-year variability of the 53 stratosphere, the magnitudes of these ozone responses may be below the threshold of 54 detectability in observations. 55 56 57 58 59 Key Words: stratosphere, ozone depletion, Hunga Tonga Hunga Ha'apai eruption, water vapor, 60 greenhouse gas 61 62 63 64 65 66

#### 67 Plain Language Summary.

Stratospheric ozone protects Earth's biosphere from harmful ultraviolet radiation, and along with water vapor, are key components in determining temperature and chemistry of the atmosphere. The January 2022 eruption of the Hunga Tonga-Hunga Ha'apai underwater volcano in the South Pacific injected water vapor into the atmosphere, increasing stratospheric water vapor by 10%. In this study, we use computer simulations of the stratosphere to project how this additional water vapor changed temperature and ozone in the months and years following the eruption. The water vapor cooled the middle stratosphere (roughly 14-25 miles above Earth's surface) and warmed the lower stratosphere (6-14 miles above the surface), with the largest changes of 2-5 degrees Fahrenheit in March-June 2022, several months after the eruption. The additional water vapor modified chemical processes that affect stratospheric ozone, leading to a projected 10-15% enhancement in the Antarctic ozone hole. This ozone hole enhancement is estimated to have maximized in October 2023, almost 2 years after the eruption, due to the slow circulation of stratospheric water vapor from the subtropics to the polar region. These impacts are expected to diminish after 2024 as the excess water vapor is slowly removed from the atmosphere by natural processes. 

#### **1. Introduction**



estimate possible future impacts over the next decade. While the HT eruption also injected a

130 modest amount of SO<sub>2</sub> which likely increased the stratospheric sulfate aerosol layer (Legras et

al., 2022; Taha et al., 2022, Zhu et al., 2022), the focus of this study is the response due only to

the water vapor injection. We quantify the projected temperature and ozone responses, as well as

the impact on various chemical constituents important to stratospheric ozone chemistry. We also

- 133
- examine the dependence of the polar ozone response on background stratospheric conditions.
- 135 136
- 137 **2. Model Simulations**
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# 139 2.1 Model description

140 Simulations in this study are conducted with the National Aeronautics and Space Administration 141 (NASA)/Goddard Space Flight Center two-dimensional model (GSFC2D), which has been used in chemistry-climate coupling studies of the stratosphere and mesosphere as well as the World 142 143 Meteorological Organization ozone assessments, including WMO (2022). The model has been 144 described and evaluated previously and has been shown to provide realistic simulations of ozone, 145 temperature, and transport-sensitive tracers for a variety of stratospheric perturbations 146 (Bacmeister et al., 1995; Jackman et al., 1996, 2016; Rosenfield et al., 1997, 2002; Fleming et 147 al., 2011; 2020). The model has very small internal variability, so that responses to small 148 perturbations can be easily detected. In Appendix A, we provide a description of recent updates 149 and model components important to the present study, including simulation of the quasi-biennial 150 oscillation (QBO). We also provide an evaluation and comparison of the model with 151 observations.

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## 153 2.2 Water vapor simulation

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155 For this study we include two water vapor tracers in the model for 2022. An unperturbed  $H_2O$ 

includes all chemical production and loss in the stratosphere and mesosphere and is specified

below the tropopause (seasonally and latitudinally dependent). In the troposphere, the 21-year

average (1981–2001) of relative humidity data from the European Center for Medium-Range

159 Weather Forecasts updated reanalysis (ERA-40) is used for the surface to 12 km, and the Upper

Atmosphere Research Satellite monthly reference atmosphere (Randel et al., 2001) is used for 12
km to the tropopause. This unperturbed H<sub>2</sub>O does not interact with other model components.

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163 To simulate the HT water vapor perturbation in the model, we use MLS version 4 (v4) data as 164 input. Previous analysis suggests that MLS v4 provides a more suitable data product to estimate 165 the HT  $H_2O$  anomaly compared to the most recent version 5 data (Millán et al., 2022). We 166 determine the zonal mean MLS water vapor anomaly in the latitude-height domain for 15 167 January 2022 through 31 December 2022 as the difference for each day from the 2005-2021 168 average for a given month. The difference of the pre-eruption 1-14 January 2022 average from 169 the 1-14 January 2005-2021 average is also removed at each latitude and altitude. This accounts 170 for the long-term trend in water vapor and is generally +0.1-0.4 ppm depending on location. The 171 MLS quality and convergence filters flagged some profiles during the first few weeks after the 172 eruption, however, the water vapor enhancements were independent of the quality and 173 convergence filtering after 8 February (Millán et al., 2022). 174 175 This daily MLS-derived H<sub>2</sub>O anomaly is added to the unperturbed model H<sub>2</sub>O at each time step 176 to create a separate perturbed  $H_2O$  tracer that interacts with the model chemistry, radiation, and 177 dynamics. This perturbed interactive H<sub>2</sub>O is updated in this manner on 15 January through 31 178 December 2022. This simulates the evolving impacts of the water vapor anomaly through the 179 end of 2022 using the MLS observations as input at each time step. Starting 1 January 2023, the 180 evolution of the perturbed H<sub>2</sub>O is fully model-computed, with the MLS-based field on 31

- 181 December 2022 serving as the initial condition.
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We also ran a parallel baseline simulation without the MLS-derived HT water vapor anomaly.
The stratospheric response to the HT H<sub>2</sub>O perturbation is taken as the difference between the
perturbation and baseline simulations.

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- 190 **3. Results**

#### **3.1** Water vapor anomaly and temperature response in 2022

193 The water vapor anomaly mixes and disperses throughout the global stratosphere during 2022, as 194 seen in the MLS-derived observations for selected months (Figure 1, top panels). While the 195 initial H<sub>2</sub>O injection at 20°S reached the upper stratosphere on 15 January (Carr et al., 2022), by 196 April the anomalous water vapor was mainly confined to the mid-stratosphere in a latitude band 197 from ~40°S to 25°N. By August and especially December, the upward tropical bulge and 198 downward midlatitude bulge of the plume reflect transport by the Brewer-Dobson circulation 199 (BDC) as noted previously in Schoeberl et al., 2022. The Southern hemisphere (SH) polar vortex 200 remained strong and isolated through much of spring 2022, confining the water vapor anomaly to 201 latitudes equatorward of ~60°S (Khaykin et al., 2022; Manney et al. 2023). The plume mixed 202 into the polar region following the vortex break-up in late November and December (Figure 1e).

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204 The corresponding model temperature response is significant and is highly correlated with the 205 H<sub>2</sub>O anomaly (Figure 1, bottom panels). The increased stratospheric water vapor enhances the IR 206 cooling above ~40 hPa (~23 km) and warming below. Maximum temperature changes occur near 207 20°S in April-May 2022, with the largest cooling of -3.2K at 20 hPa and largest warming of ~1K 208 at 54 hPa. Warming of up to several tenths of a degree K occurs around the tropical tropopause. 209 The temperature response spreads vertically and horizontally and decreases in magnitude 210 throughout 2022, following the dispersal of the water vapor plume in latitude and altitude 211 (Figure 1).

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### **3.2 Interaction with the QBO circulation**

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In this section we examine how the model temperature response to the HT H<sub>2</sub>O anomaly
compares with the NASA Modern-Era Retrospective Analysis for Research and Applications 2
reanalysis (MERRA-2, Gelaro et al., 2017). Since the meridional circulation associated with the
QBO has a significant impact in the tropical and subtropical stratosphere (Plumb and Bell, 1982),
we also examine how this circulation impacts the HT temperature response. To isolate the HT
and QBO-induced temperature responses, the long-term average seasonal cycle is removed from
MERRA-2 and the model simulations in Figures 2-4.

223	MERRA-2 assimilated temperatures show that the SH mid-stratosphere cooled substantially in
224	the months following the eruption (Coy et al., 2022). By May 2022, temperatures of nearly 5K
225	below the seasonal average occur at 20°-25°S, 20 hPa (denoted by "A" in Figure 2a). This is
226	nearly 4 standard deviations ( $\sigma$ ) colder than average at this location (denoted by "A" in Figure
227	2b), with a significant area in the mid-stratosphere at $15^{\circ}-40^{\circ}S$ greater than $2\sigma$ colder than
228	average. This contrasts with most of the stratosphere outside of the polar regions which was
229	generally within $\pm 1\sigma$ of the long-term average in May 2022 (Figure 2b). We note that
230	temperature anomalies derived from MLS version 5 data (not shown) are very similar to those
231	shown in Figure 2.
232	
233	The cooling in the mid-stratosphere is shown in the context of the MERRA-2 record since 2010
234	in Figure 3a (black line). The cooling starts shortly after the 15 Jan 2022 eruption and increases
235	for several months to a maximum of -5K ( $4\sigma$ ) in mid-May 2022. This is the second coldest
236	period during the entire 1980-2022 MERRA-2 de-seasonalized data record, surpassed only by a
237	shorter duration period of a few weeks during September 2019.
238	
239	While much of the SH mid-stratospheric cooling was caused by the HT water vapor anomaly,
240	some of the cooling was also likely due to the circulation associated with the QBO (Plumb and
241	Bell, 1982, Baldwin et al., 2001). In May 2022, the QBO was in an easterly phase below ~20
242	hPa, and the corresponding meridional circulation was characterized by sinking motion in the
243	mid-stratosphere over the equator and rising motion in the subtropics and mid-latitudes in both
244	hemispheres. This is depicted by the streamlines in Figure 2a-b which show the de-seasonalized
245	residual mean meridional and vertical winds of the Transformed Eulerian-Mean formulation
246	(e.g., Andrews et al., 1987). This circulation and the associated adiabatic heating and cooling
247	contribute to the temperature anomalies at 40-10 hPa: warm over the equator and cold anomalies
248	at $\sim 10^{\circ}$ -40° in both hemispheres.
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250	To further examine the impact of the water vapor injection and QBO circulation on these

251 observed temperature anomalies, we utilize model simulations with an interactively computed

252 QBO included (details of the QBO simulation are provided in Appendix A). Here, the model

- equatorial zonal wind is in roughly the same QBO phase as seen in the Singapore radiosonde
- observations and MERRA-2 reanalysis in May 2022, with easterlies below ~20 hPa (https://acd-
- 255 <u>ext.gsfc.nasa.gov/Data\_services/met/qbo/qbo.html#singau</u>). The corresponding model
- 256 meridional QBO circulation and associated temperature anomalies are qualitatively consistent
- 257 with MERRA-2. If the HT water vapor anomaly is not included, the SH cold anomaly is weaker
- than observed throughout the SH mid-latitudes and specifically at the location of maximum
- cooling at 20°S, 20 hPa (Figure 3a, blue line; "A" in Figure 4c). The temperature impact of the
- 260 HT water vapor anomaly in isolation shows cooling at 40-10 hPa from the SH mid-latitudes to
- the Northern Hemisphere (NH) sub-tropics and warming below ~40 hPa (Figure 4d).
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263 Including both the QBO and the water vapor anomaly brings the model into very good 264 agreement with the MERRA-2 mid-stratosphere warm and cold anomalies in the tropics and SH 265 low-mid latitudes (Figures 4a-b). The model also captures quite well the MERRA-2 extreme cold 266 anomaly at 20°S, 20 hPa following the eruption through May 2022, as well as the return to near 267 normal seasonal temperatures by the end of August 2022 (Figure 3a, red line). For the cold 268 temperature anomaly at 20°S, 20 hPa in May 2022 (~-5K), we estimate from the model that 269 ~60% (-3K) is caused by IR cooling of the excess  $H_2O$ , and ~40% (-2K) is caused by ascent of 270 the QBO-induced circulation. The model with both the QBO and H<sub>2</sub>O anomaly included also 271 qualitatively captures the warm and cold anomalies in MERRA-2 in the lower stratosphere 272 below ~40 hPa at 30°S-30°N (Figure 4a-b). These model simulations confirm that the very cold 273 mid-stratosphere anomaly in the SH subtropics in May 2022 is caused by the HT water vapor 274 anomaly but is reinforced by the ascent and adiabatic cooling caused by the meridional 275 circulation associated with the QBO being in an easterly phase in May 2022.

276

277 By August 2022, the cold temperature anomaly deepened and shifted to mid-latitudes (40°S-

278 60°S) and was prevalent throughout the SH stratosphere in the MERRA-2 data (Figure 2c-d).

279 The MERRA-2 de-seasonalized temperatures at  $40^{\circ}$ S, 27 hPa are nearly 8K (6 $\sigma$ ) colder than

- average during mid-August 2022 ("B" in Figure 2c-d, Figure 3b). This is significantly colder
- than anytime during the entire 1980-2022 MERRA-2 data record (Coy et al., 2022). However,
- the corresponding model simulation with the QBO and water vapor anomaly (Figure 3b, red line)
- captures only ~50% of the magnitude of the MERRA-2 August cold temperature anomaly.

285 We note that throughout the SH winter, the mid-upper stratosphere at mid-latitudes was 286 characterized by significantly weaker than normal wave-forced drag on the zonal mean flow, as 287 represented by the Eliassen-Palm (E-P) flux divergence due to resolved waves (Andrews et al., 288 1987) derived from the MERRA-2 reanalysis (not shown). The corresponding mid-latitude 289 residual vertical velocity field had significantly weaker than average descent at levels above  $\sim 40$ 290 hPa, as indicated by the anomalous upward motion (streamlines) in this region in August 291 (Figures 2c-d). It is therefore likely that this anomalous extratropical wave forcing contributed to 292 the extreme cold temperature anomaly observed at midlatitudes during the SH winter. However, 293 determining the source of this anomalous wave forcing is outside the scope of the 2D model used 294 in this study.

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#### **3.3 Long term water vapor and temperature response**

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298 The excess H<sub>2</sub>O is transported to the SH mid-high latitudes in the months following the eruption, 299 with values of 2-2.5 ppm reaching the SH polar region by late 2022 with the breakup of the polar 300 vortex (Figure 5a). The model simulated H<sub>2</sub>O (starting 1 January 2023) maintains concentrations 301 of ~2.5 ppm throughout the SH high latitudes in January 2023, before slowly diminishing to ~2 302 ppm by March 2023. This decrease is due to the slow removal of the excess stratospheric water 303 vapor to the troposphere as the anomaly reaches the SH mid-high latitude tropopause in early 2023. Water vapor decreases to slightly less than the baseline in early June 2023 at SH high 304 305 latitudes (Figure 5a) with the establishment of the polar vortex and onset of PSC formation. The 306 negative anomalous mixing ratios are due to the increased sedimentation of ice PSCs with the 307 enhanced water vapor relative to the baseline. Positive anomalous mixing ratios return to the SH 308 polar region in spring 2023 with the breakup of the vortex and in-mixing of mid-latitude air. 309

In the NH, the MLS-derived H<sub>2</sub>O anomaly is primarily confined equatorward of  $\sim$ 30°N until late December 2022, when increased planetary wave activity mixes the plume into the Arctic (Figure 5a). This process continues in the model simulation (starting in 2023), with anomaly values of  $\sim$ 1 ppm poleward of 60°N throughout March and April 2023.

314

- 315 The model temperature response at 25 hPa (Figure 5b) is highly correlated with the H<sub>2</sub>O plume.
- The tropics and SH cool by 0.5-3 K during 2022-early 2023, with 0.4-1K cooling in the NH
- polar region in early 2023. In the lower stratosphere at 67 hPa (Figure 5c), warming of 0.4-1K is
- 318 confined to the tropics and SH lower latitudes in 2022, with a small warming of 0.1-0.2K outside
- the polar regions ( $50^{\circ}$ S- $50^{\circ}$ N) during 2023. The water vapor and temperature anomalies
- 320 gradually diminish with time, with the H<sub>2</sub>O anomaly reduced to less than 0.1 ppm globally by
- 321 mid-2029. Mostly small cooling of 0.1-0.2K occurs throughout the stratosphere after 2023,
- 322 except in the SH polar region where larger temperature changes occur in response to the
- enhanced ozone hole. This will be discussed in section 3.5.
- 324

For 1-3 years following the eruption, the water vapor anomaly is slowly transported upwards by 325 326 the BDC, with mixing ratios of 1-1.5 ppm reaching the mesosphere by late 2023 (Figure 6a). 327 Here we show the global average since the anomalies generally have similar patterns across most 328 latitude zones (the ozone response in Figure 6c will be discussed in section 3.4). Global 329 temperature changes again follow the  $H_2O$  plume, with cooling of ~1K confined to the mid-330 stratosphere during 2022 and cooling of 1-1.5K in the mesosphere in mid-2023 through the end 331 of 2024 (Figure 6b). Small global-mean warming of 0.1-0.15K occurs in the upper 332 troposphere/lower stratosphere just after the eruption through late 2023. Starting in 2024, the 333 global average temperature response is almost exclusively negative (cooling) throughout the 334 middle atmosphere as the anomalies gradually diminish to be quite small by 2030. 335 336 The total global burden (mass) of the  $H_2O$  anomaly is 150-160 Tg shortly after the eruption 337 through mid-2023 and is projected to decrease thereafter (Figure 7). The excess stratospheric 338 water vapor is slowly removed by sedimentation of PSCs within the Antarctic vortex, as well as 339 return of stratospheric air to the troposphere by the BDC at mid-high latitudes of both 340 hemispheres. The combination of these processes leads to an exponential decay of the anomaly, 341 with an estimated average e-folding time of 2.5 years from mid-2023 through 2031 (Figure 7, red 342 dashed-dotted line). The burden is reduced to 4.7 Tg by the end of 2031, which is ~3% of its 343 starting value in January 2022.

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#### 345 **3.4** Global profile ozone and related chemical responses

347 Ozone is affected by the HT water vapor anomaly globally throughout the middle atmosphere 348 due to changes in photochemistry, both directly, and indirectly via changes in the temperature-349 dependent ozone loss cycles (e.g., Dvortsov and Solomon, 2001; Brasseur and Solomon, 2005). 350 MLS observations show that ozone in the mid stratosphere decreased significantly in the SH 351 subtropics and midlatitudes starting in early winter 2022 (Wang et al., 2022). However, our 352 model simulations suggest that the ozone response to the HT H<sub>2</sub>O anomaly in this region is quite 353 small in 2022. This is discussed further in section A.4.3 of Appendix A, along with comparisons 354 to the MLS data.

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356 For globally averaged ozone, maximum projected changes occur in 2023-2024, ~1-2 years after 357 the eruption (Figure 6c). During this time, the  $H_2O$  anomaly is 20-25% above the background in 358 the SH mid-stratosphere, and ~15% above the background globally throughout the mesosphere 359 (Figure 8a). One of the direct consequences of excess H<sub>2</sub>O is the increase in odd hydrogen 360 species (HOx =  $H+OH+HO_2+2*H_2O_2$ ), with the OH radical increasing by 5-10% throughout the 361 stratosphere and mesosphere in 2023-2024 (Figure 8b). This enhances the HOx catalytic loss 362 cycle, which is the major contributor to the total odd oxygen  $(O+O_3)$  chemical loss above ~50 363 km (Figure 9b). The resulting projected global ozone loss is >0.5% at altitudes above ~40 km 364 throughout 2023-2026 (Figure 6c) and increases with altitude to 5-10% above ~60 km from mid-365 2023 to mid-2025.

366

367 Changes in the total odd nitrogen family

368 (NOy=N+NO+NO<sub>2</sub>+NO<sub>3</sub>+2\*N<sub>2</sub>O<sub>5</sub>+HNO<sub>3</sub>+HO<sub>2</sub>NO<sub>2</sub>+HONO+ClONO<sub>2</sub>+BrONO<sub>2</sub>; Figure 9a,

369 orange line) and a subset of NOy directly involved in the odd nitrogen-ozone loss cycle

370  $(NOx=N+NO+NO_2+NO_3+2*N_2O_5;$  Figure 9a, blue line) are mostly negative throughout the

371 stratosphere and mesosphere. Decreases in NOx are 5-10% in the SH polar lower stratosphere

- 372 due mainly to increased sedimentation of nitric acid tri-hydrate (NAT) PSCs in the Antarctic
- 373 vortex (e.g., Toon et al., 1986), with smaller NOx decreases of 2-4% in the Arctic (Figure 8d).

374 Decreases in global NOx maximize in the mid-stratosphere mainly due to increased OH which

375 converts NOx to HNO<sub>3</sub> via the OH+NO<sub>2</sub> reaction (Figure 9a). There is also a small contribution

to this NOx decrease due to a slight increase in the heterogeneous reaction

377 N<sub>2</sub>O<sub>5</sub>+H<sub>2</sub>O $\rightarrow$ 2\*HNO3 on sulfates. Although the model stratospheric sulfate aerosol surface area 378 is specified and does not interact with the H<sub>2</sub>O anomaly, the total rate of this reaction is slightly

faster (1-3%) at 20-30 km at mid-high latitudes due to the increased water vapor.

380

381 In the mesosphere where the HNO<sub>3</sub> concentration is very small and NOx  $\approx$  NOy, odd nitrogen

decreases by 2-5% mainly due to the colder temperatures (Figures 8d, 9a). Here, the abundance of atomic nitrogen (N) is increased due to the reduced rate of the strongly temperature dependent reaction  $N+O_2 \rightarrow NO+O$  at lower temperatures. The increased N increases the loss of NOy which is controlled by the reaction  $N+NO \rightarrow N_2+O$  (Rosenfield and Douglass, 1998).

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387 The model ozone changes in the mid-stratosphere (30-5 hPa, ~25-37 km) are projected to be 388 predominantly positive, starting shortly after the eruption in early 2022 and lasting through 2029 389 (Figures 6c). Maximum global ozone increases of 0.5-1% occur in mid-2022 through the end of 390 2024. In this region, the NOx catalytic cycle dominates the total odd oxygen chemical loss. 391 Because of reduced NOx, anomalous NOx-odd oxygen loss is positive throughout the 392 stratosphere (Figure 9b, blue line), and the dominance of this loss cycle in the mid-stratosphere 393 leads to a small positive total odd oxygen chemical tendency (Figure 9b, black line) and positive 394 ozone change in this region (Figure 8e). This result is qualitatively consistent with previous 395 studies that found reduced mid-stratospheric NOx-ozone loss due to increased HOx 396 concentrations from methane oxidation (Nevison et al., 1999; Randeniya et al., 2002). 397

In the lower stratosphere below ~23 km (40 hPa) the ozone response is projected to be mostly
negative (Figures 6c and 8e), with the global response mainly reflecting a deepened Antarctic
ozone hole. This will be discussed in the next section.

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#### 403 3.5 Antarctic profile ozone and related chemical responses

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405 MLS data show that lower stratospheric Antarctic ozone during spring 2022 was significantly

406 lower than the 2005-2021 average (Figure 10d). Previous model results show that this low ozone

407 could be explained by the HT aerosol perturbation combined with colder than average

- 408 temperatures in the polar vortex (Wang et al., 2022). For the model simulations presented in this
- 409 study, the SH planetary wave forcing (section A.2) was reduced during winter-spring 2022 to
- 410 mimic the strong and isolated SH polar vortex that persisted well into November. However, the
- 411 resulting additional ozone depletion due to the HT H<sub>2</sub>O anomaly was significantly smaller than
- 412 shown in the de-seasonalized MLS data in spring 2022 (Figure 10c-d). The model shows some
- 413 qualitative consistency with the MLS ozone during the first half of 2023, with a negative
- 414 anomaly at 10-20 km and a positive anomaly at 20-25 km.
- 415

In late winter-spring 2023, the model simulates a deepened ozone hole, with a significant

417 negative anomaly at ~10-25 km which is qualitatively consistent with MLS at most altitudes

418 (Figure 10c-d). However, the persistence of a positive anomaly seen in MLS in a shallow layer

419 near 20 km through mid-October 2023 is not captured in the model. Some of these differences

420 are likely due to the background stratospheric variability which makes it difficult to detect the

421 impact of the excess H<sub>2</sub>O in the de-seasonalized MLS ozone data. Model biases, at least for the

- 422 first 1-1<sup>1</sup>/<sub>2</sub> years after the eruption, also may be due in part to not including the HT aerosol
- 423 perturbation in the model.
- 424

425 The processes that cause the ozone hole have been well established in past studies (e.g., 426 Solomon, 1999; Solomon et al., 2014, Solomon et al., 2015). To examine in more detail how the 427 excess H<sub>2</sub>O impacts the model ozone hole, we focus on lower stratospheric anomalies for April-428 December 2023 of several constituents relevant to the chemistry driving the enhanced ozone 429 hole. By mid-winter 2023, the water vapor anomaly is present within the Antarctic vortex 430 throughout the depth of the stratosphere with corresponding anomalous cooling (Figures 10a-b). 431 The combination of the H<sub>2</sub>O and temperature anomalies leads to projected: 1) increase in type I 432 NAT PSCs in early winter with a corresponding decrease in gas-phase HNO<sub>3</sub>, and 2) increase in 433 type II ice PSCs throughout the winter (Figures 11a-b). The increased sedimentation of NAT 434 PSCs causes denitrification (e.g., Toon et al., 1986) starting in early June, with negative 435 anomalous NOy persisting through winter and spring and negative anomalous NOx 436 concentrations in October-November (Figure 11b).

437

438 The projected enhancement in PSCs and PSC surface area increases the heterogeneous 439 conversion of chlorine and bromine from reservoir species (HCl, ClONO<sub>2</sub>, HOBr, BrONO<sub>2</sub>) to 440 reactive forms that destroy ozone (Cl, ClO, Br, BrO). The anomalous heterogeneous chlorine 441 activation on sulfate aerosols is also generally faster due to the lower temperatures and increased 442 water vapor (e.g., Solomon, 1999; Burkholder et al., 2019), even though the model sulfate 443 aerosol surface area is specified and does not interact with the H<sub>2</sub>O anomaly. The anomalous rate 444 of a key heterogeneous reaction,  $ClONO_2 + HCl \rightarrow HNO_3 + Cl_2$  on PSCs and sulfates, is shown in Figure 11b (purple dotted line). The anomaly maximizes in June following the anomalies in 445 446 PSC surface area and enhancement of the reaction on sulfates. The secondary anomaly maximum 447 in September-October is caused by an increase in the sulfate reaction due to lower temperatures (Figure 11e), as the ice and NAT PSC and H<sub>2</sub>O anomalies are all quite small in the spring. 448 449 Increased conversion of chlorine to reactive forms on sulfate aerosols under cold SH polar conditions was noted previously (e.g., Hanson et al., 1994). 450

451

452 The chemical loss of odd oxygen due to the chlorine and bromine catalytic cycles is enhanced 453 with increased sunlight in early August through late October, and this controls the total chemical 454 loss (Figures 11c-d). The additional ozone depletion (Figure 11d, black line) drives a significant 455 reduction in the solar ultraviolet ozone heating and temperature starting in mid-late August as the 456 solar elevation increases (Figure 11e). This further enhances the chemical ozone loss and 457 reduction in heating, leading to a delay in the breakup of the polar vortex. This delays the 458 increase of odd oxygen due to transport into the polar region associated with the vortex breakup 459 (Figure 11d, green dashed-dotted line), which in turn further enhances the reduction of polar 460 ozone prior to the vortex breakup. We note that the lower stratospheric Antarctic spring cold 461 temperature anomaly occurs yearly throughout 2022-2029 (Figure 5c).

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Anomalous ozone concentrations at 54 hPa reach a minimum of ~-5 DU/km in late October
(Figure 11d, black line), before recovering to near-baseline values by mid-December. The
recovery is driven mainly by transport associated with the vortex breakup as indicated by the
transport tendency (Figure 11d, green dashed-dotted line), with a smaller contribution due to
reduced chemical loss as indicated by the positive total chemical tendency (Figure 11d, magenta
line). This positive chemical tendency is due to the reduced anomalous NOx concentrations and

a decrease in the NOx catalytic loss cycle in October-November (Figure 11b-c, blue lines). NOy
and NOx return to near-baseline concentrations in mid-December following the vortex breakup
and in-mixing of NOy-rich midlatitude air.

472

473 In the middle stratosphere (~40-10 hPa), positive model ozone anomalies in the Antarctic occur 474 from December 2022-June 2023 (Figure 10c), mainly due to the reduced NOx catalytic loss 475 cycle (i.e., increased odd oxygen tendency, Figure 12 blue line). A negative ozone anomaly 476 driven by the increased halogen loss cycles occurs during August-September 2023. Relatively 477 small ozone changes occur during late September-October as the positive transport tendency 478 largely offsets the anomalous chemical ozone loss. Positive ozone anomalies re-emerge in the 479 mid-stratosphere during December 2023 through June 2024 (Figure 10c), driven by a 480 combination of reduced NOx loss and positive anomalous transport tendency. This anomalous 481 polar transport occurs during the SH late spring-summer and is a dynamical response to the 482 enhanced ozone hole. This feature has been discussed in previous modeling studies (Kiehl et al., 483 1988; Mahlman et al., 1994; Stolarski et al., 2006), and has been seen in observations of 484 temperature (Randel and Wu, 1999) and ozone (Stolarski et al., 2006). For the HT H<sub>2</sub>O response, 485 the additional ozone depletion leads to a projected delay in 1) the spring vortex breakup, and 2) 486 the corresponding wave-forced drag on the zonal mean flow and acceleration of the BDC. As a 487 result, there is anomalous descent in the Antarctic mid-upper stratosphere and positive odd 488 oxygen transport tendency relative to the baseline in late November-December 2023 (Figure 12). 489 The anomalous descent and associated adiabatic warming cause the positive temperature 490 anomaly in the polar mid-stratosphere above ~40hPa during November-December 2023 (Figure 491 10b). This warm anomaly is a yearly recurring feature in the Antarctic summer mid-stratosphere 492 throughout 2023-2030 (Figure 5b).

493

In the very lower stratosphere below ~70 hPa (~18 km), small negative ozone anomalies persist
through summer and fall 2024 (Figure 10c) as the transport processes associated with the delayed
vortex breakup do not return ozone quite to the baseline value at these altitudes.

- 497
- 498

#### 499 **3.6 Total ozone response**

### 501 3.6.1 Baseline response

502

503 The additional ozone loss due to the HT water vapor injection results in a projected 25-30 DU 504 enhancement of the Antarctic ozone hole during spring 2023 (Figure 13). The response 505 diminishes to 20-25 DU in spring 2024, with decreasing severity in the ozone hole enhancement 506 thereafter as the water vapor perturbation diminishes. In 2022, a strong and isolated SH polar 507 vortex persisted well into November, with large observed ozone losses and a deeper than normal 508 ozone hole relative to the historical data record (e.g., Newman and Lait, 2023). However as 509 discussed in section 3.5, the H<sub>2</sub>O anomaly resulted in a relatively small model ozone hole 510 enhancement of 5-7 DU in spring 2022 (Figure 13). This springtime ozone loss was limited since 511 the additional H<sub>2</sub>O was confined to the SH mid-stratosphere during mid-winter and did not reach 512 the polar lower stratosphere until late November 2022 (Figure 10a; see also Manney et al., 2023) following the polar vortex breakup, after the time of normal seasonal formation of PSCs. 513 514 515 In the Arctic, the anomalous water vapor and stratospheric cooling cause additional springtime 516 ozone depletion due to processes that are generally like those in the Antarctic as discussed above. 517 However, the projected Arctic ozone losses are much smaller, 4-5 DU, and do not start until 518 spring 2024 (Figure 13). The anomalous Arctic ozone depletion diminishes with time to be < 1

519 DU by the late 2020s. At mid-latitudes, maximum total ozone decreases are 4-6 DU in the SH

and 1-2 DU in the NH, with changes of less than  $\pm 1$  DU in the tropics throughout the post-

- 521 eruption period.
- 522

523 3.6.2 Sensitivity to stratospheric background conditions

524

525 The projected ozone responses shown in Figures 13 are from a model simulation that uses the 526 standard planetary wave forcing which gives climatologically averaged stratospheric conditions 527 (section A.2). This simulation is generally consistent with the observed long term average 528 seasonal cycle in polar total ozone in each hemisphere (Figure 14, orange vs. black solid lines). 529 However, the polar regions are characterized by large interannual variability in winter and spring 530 caused primarily by variations in planetary wave driving in the stratosphere. This is depicted by the gray shading in Figure 14 which shows the range in historical total ozone observations for1991-2022 (Newman and Lait, 2023).

533

534 To examine the dependence of the ozone response on the background stratospheric conditions, 535 we ran a series of experiments with the stratospheric wave driving varied to mimic the observed 536 range in total ozone shown in Figure 14. Here we focus on the model year with the largest 537 anomalous ozone loss in each polar region: 2023 for the SH and July 2023-June 2024 for the 538 NH. Substantially increased planetary wave forcing gives warm polar stratospheric conditions 539 and total ozone at the upper end of the range in the historical data record in both hemispheres 540 (Figure 14, red solid lines). Conversely, substantially reduced wave forcing results in a cold polar stratosphere and total ozone at the lower end of the data record (blue solid lines). For each 541 542 wave driving case, the water vapor anomaly simulation is depicted by the dashed lines in Figure 543 14.

544

545 In the SH, including the HT H<sub>2</sub>O perturbation under the strong wave forcing (warm) conditions 546 results in somewhat less projected ozone depletion compared to the standard wave forcing, with 547 polar cap average additional depletion of 13 DU vs. 20 DU in 2023 (Figure 14a). The stronger 548 wave driving, and warmer stratospheric conditions also promote a faster return to the baseline 549 total ozone in December 2023 compared to the standard case. Under the weak wave forcing 550 (cold) conditions, the baseline already has very low ozone concentrations in the lower 551 stratosphere, so that including the H<sub>2</sub>O anomaly has less of an impact than with the standard 552 wave forcing, with additional polar cap average depletion of 14 DU (Figure 14a, blue dashed 553 line). The largest impact occurs under conditions slightly warmer than the standard case (not 554 shown). Here, the larger ozone concentrations available in the baseline combined with 555 substantial anomalous chlorine and bromine activation resulted in additional polar cap depletion 556 of 23 DU, compared to 20 DU for the standard wave forcing case.

557

558 In the Arctic, the projected model ozone response to the additional water vapor has a small

dependence on the background stratospheric conditions in spring 2024 (Figure 14b). The colder

560 conditions with weak wave driving result in 5 DU additional polar cap average depletion

561 compared to the standard (3 DU) and strong (3 DU) wave driving cases. However, even under

562 the cold conditions of the weak wave driving case, the additional model ozone depletion in the 563 Arctic is small compared to the Antarctic. This is due to a combination of factors, one being the 564 smaller anomalous H<sub>2</sub>O concentrations transported to high NH latitudes. Just prior to the onset of 565 PSC formation in early winter (November-December 2023), the excess water vapor is 0.2-1 ppm 566 in the Arctic lower stratosphere, substantially less than the 1-2 ppm in the early winter Antarctic 567 (May-June 2023). Another important factor is the generally warmer Arctic temperatures that 568 limit additional ozone depletion (Solomon et al., 2014). For example, in the weak wave driving 569 (cold) case, model Arctic temperatures throughout the lower stratosphere in February-April 2024 570 are still 3-7K warmer compared to the Antarctic strong wave driving (warm) case in late winter – 571 spring 2023. We note that these model NH ozone responses to the H<sub>2</sub>O anomaly are generally 572 consistent with previous studies of the Arctic ozone response to stratospheric water vapor 573 changes using 3-D chemical transport models driven by meteorological reanalysis (e.g., Vogel et 574 al., 2011; Thölix et al., 2018).

575

576 Figure 14 suggests that the total ozone impact of the HT H<sub>2</sub>O anomaly is generally smaller than 577 the year-to-year variability characteristic of the stratospheric polar regions during winter and 578 spring. The standard deviation of the observed SH October-November polar cap total ozone is 579 ~35 DU during 1991-2022, significantly larger than the maximum model estimated response to 580 the H<sub>2</sub>O anomaly of 23 DU. The difference is even larger in the Arctic spring, with a March-581 April observed standard deviation of ~26 DU compared to a maximum model anomaly response 582 of 5 DU. Therefore, it is possible that the response to the HT water vapor injection may not be 583 easily detectable above the background variability in observational total ozone data.

- 584
- 585

## 586 **4. Summary and Conclusions**

587

588 The January 2022 Hunga Tonga-Hunga Ha'apai volcanic eruption increased stratospheric water

589 vapor by ~10% (~150 Tg) (Millán et al., 2022) which significantly altered the radiative balance,

dynamics, and photochemistry of the stratosphere (Vömel et al., 2022; Coy et al., 2022;

591 Schoeberl et al., 2022, Sellitto et al., 2022; Schoeberl et al., 2023). In this study, we examine

how this unique natural perturbation impacted stratospheric temperature and ozone in the first 12 years following the eruption and estimate possible future responses over the next decade.

594

595 The maximum radiatively-induced model temperature response occurs in March-June 2022, 596 several months after the eruption, with a cooling of 2-3K in the SH mid-stratosphere, and ~1K 597 warming in the lower stratosphere. This radiatively-driven warming is as much as several tenths 598 of a degree K around the tropical tropopause, which may have important implications for the 599 amount of water vapor entering the stratosphere. However, quantification of this effect is beyond 600 the scope of the 2D model used in this study. We note that these impacts do not include sea 601 surface temperature feedback, which may be important to fully quantify the response to the 602 radiative forcing of the HT H<sub>2</sub>O anomaly.

603

The QBO was in an easterly phase in April-May 2022, and model simulations suggest that ascent and adiabatic cooling associated with the QBO circulation, combined with the radiative cooling of the H<sub>2</sub>O anomaly, can explain the near-record cold temperatures seen in the MERRA-2 reanalysis in the SH subtropical mid-stratosphere during May 2022.

608

609 Transport of the water vapor plume to the Antarctic lower stratosphere is delayed until late 2022 610 with the breakup of the polar vortex (Manney et al., 2023). The plume reaches the Arctic in 611 winter 2022-2023 and is slowly transported to the mesosphere during 2023-2024 by the rising 612 branch of the Brewer-Dobson circulation. Radiatively-induced temperature changes are projected 613 to be small in the NH stratosphere ( $<\pm 0.5$ K), with larger cooling in the mesosphere of 1-1.5K 614 that peaks in late 2023-2024. The increased water vapor and cooling in the mesosphere may have 615 implications for polar mesospheric cloud formation (e.g., Hervig et al., 2016; Lübken et al., 616 2018).

617

The anomalous water vapor impacts the chemistry in the middle atmosphere by increasing OH concentrations. This increases the odd hydrogen-ozone loss cycle, which is dominant in the mesosphere, causing a projected 5-10% reduction in mesospheric ozone globally from mid-2023 to mid-2025. The additional OH also increases the conversion of odd nitrogen to HNO<sub>3</sub>, thereby reducing the NOx-ozone loss cycle throughout the stratosphere. This results in a small net global ozone increase of 0.5-1% during 2023-2024 in the mid-stratosphere where the NOx catalyticcycle dominates the chemical loss of odd oxygen.

625

626 In the lower stratosphere, the additional water vapor is projected to increase PSC surface area 627 and the heterogeneous conversion of chlorine and bromine into reactive forms that destroy 628 ozone. This causes a deepened ozone hole in the Antarctic spring. The decrease in solar ozone 629 heating reduces temperatures and delays the break-up of the vortex, further enhancing the ozone 630 loss. This effect is projected to maximize in 2023 with 25-30 DU enhancement of the ozone hole. 631 Model sensitivity simulations suggest that this response will be somewhat dependent on the 632 background stratospheric conditions, with an estimated polar cap average range of 13-23 DU 633 additional ozone depletion. In the Arctic, additional ozone losses due to the excess water vapor 634 are relatively small, with an estimated maximum additional depletion of 3-5 DU in spring 2024 635 and slightly smaller losses of 2-4 DU in spring 2025 and 2026. The Arctic responses have a 636 relatively small dependence on the background stratospheric conditions.

637

By mid-2023 and beyond, the excess  $H_2O$  is slowly removed by return to the troposphere at midhigh latitudes and by sedimentation of PSCs within the Antarctic vortex. The anomaly decays exponentially with a projected e-folding time of 2.5 years, and the corresponding temperature and ozone responses diminish slowly after 2024. By the end of 2031, the additional  $H_2O$  is estimated to be ~3% of its initial value of ~150 Tg, with very small temperature and ozone responses.

644

645 The focus of the present paper is on the response due only to the HT  $H_2O$  anomaly in isolation so 646 that sulfur impacts are not considered. The HT eruption injected a modest amount of SO<sub>2</sub> (0.4-647 0.5 Tg) which likely increased the stratospheric sulfate aerosol layer (Legras et al., 2022; Taha et 648 al., 2022; Zhu et al., 2022). This could impact ozone chemistry via increased heterogeneous 649 conversion of odd nitrogen to  $HNO_3$ , and increased chlorine activation at lower temperatures 650 (~195K). Stratospheric temperatures can also be affected via changes in ozone and aerosol 651 absorption of infrared radiation. These impacts can occur globally and in the polar regions as 652 shown for previous volcanic eruptions (e.g., Brasseur and Granier, 1992; Randel et al., 1995; 653 Solomon et al., 1996; Portmann et al., 1996). For the HT eruption, Wang et al. (2022) showed

654 that significant additional ozone depletion occurred in the Antarctic lower stratosphere in spring 655 2022 when including the  $SO_2$  injection in model simulations. Therefore, the ozone response to 656 the H<sub>2</sub>O injection presented here likely somewhat underestimates the full response to the HT 657 eruption, at least in the first year following the eruption. 658 659 The model-projected ozone responses presented here are generally smaller than the natural 660 variability of the stratosphere. The largest response is likely the enhancement of the Antarctic 661 ozone hole. However, the additional model ozone loss is at most 23 DU (2023 polar cap 662 average), which is smaller than the standard deviation of springtime polar total ozone 663 observations in the SH (~35 DU). Therefore, it is possible that any ozone response to the HT 664 water vapor injection may not be detectable above the background variability in observational 665 data sets. 666 667 668 **Appendix A: GSFC2D Model Description and Evaluation** 669 670 671 In this appendix, we provide a description and evaluation of the GSFC2D model, focusing on 672 recent updates and model components important to the present study. We also provide some 673 model evaluation via comparisons with observations of age of air, H<sub>2</sub>O, and ozone. 674 675 A.1 Model chemistry and radiation 676 677 The model has full stratospheric chemistry, with a diurnal cycle computed for all constituents 678 each day. Transport of all constituents follows the chemistry calculations at each time step within 679 the diurnal cycle. This is updated from the previous scheme in which only the diurnal averages 680 were transported at the end of the diurnal cycle. The resulting changes are most significant at 681 mid-high latitudes during late autumn through early spring when the photochemical time scales 682 are long, and the new methodology improves the simulations of polar ozone compared with 683 observations (see Figure 14). The model domain extends from the surface to ~92 km (.002 hPa) with a grid spacing of 4° latitude and 1 km in altitude. 684

Time dependent surface mixing ratio boundary conditions are taken from WMO (2022) for the major ozone depleting substances and Meinshausen et al. (2020) for the major greenhouse gases  $CO_2$ ,  $CH_4$ , and  $N_2O$ . The latest JPL-2019 recommendations (Burkholder et al., 2019) are used for the kinetic reaction rates, photolysis cross sections, and heterogeneous reactions on the surfaces of polar stratospheric clouds (PSCs) and stratospheric sulfate aerosols. The stratospheric aerosol surface area density is specified from the CMIP6 dataset based on Kovilakam et al. (2020).

692

693 A parameterization to simulate type Ia (solid nitric acid trihydrate, NAT) and type II (Ice) PSC 694 formation follows Considine et al. (1994) and includes sedimentation of NAT and ice aerosols. 695 Calculations of PSC occurrence frequency and surface area density use a combination of the 696 zonal mean temperatures computed in the model and longitudinal temperature probability 697 distributions (deviations from the zonal mean, T') obtained from the MERRA-2 reanalysis. Here, 698 the MERRA-2 climatological T' distribution is based on daily data averaged over 2000-2020 and 699 is added to the model-computed zonal mean temperatures at each time step. This hybrid 700 methodology allows the PSCs to respond to both observational-based longitudinal variations and 701 the evolving model zonal mean temperature. To convert the PSC concentration to surface area 702 density, a lognormal particle size distribution is assumed (Considine et al., 1994). The uptake 703 coefficients for heterogeneous reactions on PSCs are specified to be constant (Burkholder et al., 704 2019). We note that supercooled ternary solution (STS, H<sub>2</sub>SO<sub>4</sub>/HNO<sub>3</sub>/H<sub>2</sub>O) type Ib PSCs are not 705 computed in the current model configuration.

706

Effects of spherical geometry in the photolysis and solar heating rate calculations are
approximated by use of the Chapman function (McCartney, 1976), accounting for twilight
conditions for solar zenith angles up to 94°. For the infrared (IR) parameterization, the model
uses the Rapid Radiative Transfer Model for GCM Applications (RRTMG), a state-of-the-art
algorithm that is used in various climate and weather forecast models (Mlawer et al., 1997;
Clough et al., 2005; Hurwitz et al., 2015).

713

A.2 Model dynamical parameterizations

715

The planetary wave parameterization (Bacmeister et al., 1995; Fleming et al., 2011) uses lower
boundary conditions at 750 hPa (~2 km) of geopotential height amplitude and phase for zonal
wave numbers 1-4. These are derived as a function of latitude and season using a 30-year
average (1991-2020) of MERRA-2 data for the standard model wave forcing. These boundary
conditions can be adjusted to modify the planetary wave forcing in the stratosphere, thereby

721 giving colder or warmer than average conditions during the winter and spring in each

hemisphere. In section 3.6.2, we examine the sensitivity of the HT  $H_2O$  anomaly on the

723 background stratospheric conditions using this modified wave forcing.

724

725 Momentum deposition and vertical eddy diffusion from breaking gravity waves in the 726 stratosphere and mesosphere are computed following the parameterization originally developed 727 by Lindzen (1981) and modified by Holton and Zhu (1984). The parameterization solves for a 728 general spectrum of monochromatic waves with phase speeds covering the range of  $\pm 40$  m/sec at 729 intervals of 10 m/sec. The momentum flux is specified in the upper troposphere (~325 hPa) as a 730 function of latitude and season, with the slower phase speed waves having larger momentum flux 731 (Holton and Zhu, 1984). The parameterization also solves separately for a single stationary 732 gravity wave generated by flow over orography. Here, vertical profiles of momentum deposition are computed on a longitude-latitude grid ( $10^\circ \times 4^\circ$  grid spacing), with the model zonal mean 733 734 zonal wind used for each longitude. The orographic surface forcing is based on a multi-year 735 average of monthly zonal gravity wave surface stress from the earlier version 3 of the Whole 736 Atmosphere Community Climate Model (Garcia et al., 2007; see also McFarlane, 1987).

737

The zonally averaged momentum deposition from gravity waves and planetary waves is used in
the 2D model zonal wind and meridional circulation calculations. The resulting model zonal
wind and temperature distributions compare well with multi-year averaged monthly
meteorological reanalysis (e.g., Fleming et al., 2011).

742

743 A.3 Model QBO simulation

744

Simulation of the QBO in equatorial zonal wind is based on previous 2D model studies whichparameterize the momentum deposition from thermally damped large scale, long period Kelvin

and Rossby-gravity waves (e.g., Plumb and Bell, 1982; Gray and Pyle, 1989; Dunkerton, 1997).

- 748 We include two slow Kelvin waves (zonal wavenumber 2) with phase speeds of +20 m/sec and
- +30 m/sec, and a Rossby-gravity wave (zonal wavenumber 4) with a phase speed of -40 m/sec. A
- fast Kelvin wave with phase speed of +60 m/sec is also included to simulate the westerly phase
- 751 of the semiannual oscillation in the upper stratosphere and lower mesosphere.
- 752

Previous work has shown the importance of including the momentum flux from small scale gravity waves in generating a realistic QBO (Dunkerton, 1997; Geller et al., 2016). We use the parameterization described in section A.2 with high vertical resolution (250 meters) to compute the momentum flux from a spectrum of equatorial gravity waves with phase speeds covering the range of  $\pm 40$  m/sec at intervals of 2 m/sec (41 waves).

758

759 For each large scale and gravity wave component, the momentum flux is specified at the bottom 760 boundary in the upper troposphere ( $\sim$ 325 hPa), with the slower phase speed gravity waves 761 having larger momentum flux (Dunkerton, 1997). As discussed in previous studies (Geller et al., 762 2016), the input parameters of wave phase speed and lower boundary momentum flux, 763 respectively, are adjusted to obtain a QBO with realistic amplitude and period. The model 764 equatorial zonal wind shows good agreement with Singapore radiosonde data and the MERRA-2 reanalysis (Coy et al., 2016) in reproducing the general features of the QBO, including a period 765 766 of ~28 months and similar rate of downward phase progression. This is shown in a representative 767 comparison with MERRA-2 in Figure A1. Note that the model is free running and does not 768 correspond to a specific year as is represented in the reanalysis. The maximum model zonal wind 769 QBO amplitude of ~20 m/sec occurs at 10-30 hPa, decreases to ~5-6 m/sec at 70 hPa, and is near 770 zero at the tropical tropopause.

771

Momentum forcing from the different wave components is specified to decrease rapidly away from the equator, with a latitudinal dependence for the large-scale waves as in Gray and Pyle (1989). The resulting model QBO amplitude has a latitudinal variation consistent with observations (Wallace, 1973), with a half width of 10-15 degrees. The meridional circulation associated with the QBO (Plumb and Bell, 1982) is also consistent with observations, as seen in the circulation-induced temperature changes over the equator and in the subtropics discussed in section 3.2 (Figure 4). This circulation is also important for tracer transport in this region (Trepte
and Hitchman, 1992; Randel et al., 1998; Baldwin et al., 2001).

780 781

782 A.4 Comparison of model tracers with observations

783

In this section, we provide an evaluation of the model transport fields by comparing the age of
air and H<sub>2</sub>O simulations with observations. We also show the model ozone response to the HT
H<sub>2</sub>O anomaly compared with MLS observations at SH low-middle latitudes during 2022-2023.

788 A.4.1 Age of Air

789

Stratospheric mean age of air is a widely used diagnostic that tests the overall fidelity of model transport. Figure A2 shows the age of air derived from measurements of SF<sub>6</sub> and CO<sub>2</sub> at 20 km (~50 hPa) and vertical profiles for three latitudes zones during the 1990s (Hall et al., 1999), along with the model simulation. There are differences in the observations at middle and higher latitudes that may reflect photochemical influences on SF<sub>6</sub> which would cause an overestimation in the inferred ages (Hall and Waugh, 1998). Some of the older age measurements at 65°N may also reflect remnants of the polar vortex (Ray et al., 1999).

797

For the most part GSFC2D compares generally well with the observations in reproducing the absolute values and the latitudinal and vertical gradients. The model slightly underestimates the observations at 20 km at 30°N-45°N, and at NH high latitudes above 30 km. However, the good overall agreement illustrates that the model stratospheric transport rates, including the relative magnitudes of vertical motion and horizontal mixing, are generally realistic. This is also important to provide a reasonable simulation of the H<sub>2</sub>O anomaly decay rate (Figure 7).

804

A.4.2 Water vapor

806

Here we compare the full water vapor field (background plus HT anomaly) for 2022-2023 from a
simulation in which the model is forced with the MLS-derived H<sub>2</sub>O anomaly (section 2.2), but

only through the end of February 2022. Starting 1 March 2022, evolution of the full H<sub>2</sub>O field is
model computed.

811

812 The model simulates the full water vapor field generally well in the latitude-height domain 813 compared with MLS version 5 (v5, Figure A3), as well as the time evolution through November 814 2023 in the mid-stratosphere (Figure A4). The model reproduces the observed transport of large 815 H<sub>2</sub>O concentrations (> 7 ppm) associated with the HT anomaly from the initial injection to SH 816 mid-high latitudes through November 2023. The  $H_2O$  anomaly appears in the Arctic as a small 817 enhancement in the MLS data starting in early 2023, and this feature is qualitatively similar in 818 the model. The model underestimates the MLS observations by ~0.5 ppm throughout much of 819 the stratosphere, likely reflecting the amount of H<sub>2</sub>O entering through the tropical tropopause in 820 the model which is specified from the Upper Atmosphere Research Satellite monthly reference 821 atmosphere (Randel et al., 2001). Some of the model differences with MLS are also due to 822 atmospheric variability not resolved in the simulation. Isolation of the tropical stratosphere 823 below ~20 hPa tends to be somewhat overestimated in the model, as indicated by the stronger 824 latitudinal gradients at 15°S-15°N compared with MLS, especially during 2022 (Figure A4). 825

The model simulates the Antarctic H<sub>2</sub>O distribution generally well compared with MLS (Figure A5), although the model has a small high bias in the late winter-spring below ~23 km. Some of this may be due to interannual variability and the model not fully resolving the observed isolation of the SH polar region through late spring 2022 (Manney et al., 2023) and through

830 September 2023, as well as possible underestimation of ice sedimentation.

831

832 Both the MLS data and model show a return to more typical low H<sub>2</sub>O values (< 4-5 ppm) in the 833 equatorial mid stratosphere by early 2023 (Figure A4). This reflects the isolation of the tropics 834 from mid-latitudes and the upward transport of drier air from the tropopause, as seen in the 835 equatorial time-height sections (Figure A6a-b). As discussed in section 3.2, the model zonal 836 wind QBO in Figure A6b is in roughly the same phase as the Singapore radiosonde data, with 837 easterlies at 30 hPa at the time of the eruption through mid-2022 ("E" in Figure A6a-b). The 838 corresponding QBO-induced circulation, with relative descending motion over the equator, slows 839 the overall ascent in the tropical mid-stratosphere. As a result, upward transport of the enhanced

H<sub>2</sub>O is relatively slow throughout most of 2022, and the model compares well with the MLS data

- in this regard (Figure A6a-b). By late 2022 early 2023, the QBO is in a westerly phase ("W" in
- Figure A6a-b) so that the QBO-induced circulation enhances the overall ascent in the tropics.
- 843 This results in more rapid upward transport of the H<sub>2</sub>O plume with large water vapor
- concentrations (> 7 ppm) in the equatorial upper stratosphere through November 2023 in both

845 MLS and the model.

- 846
- 847 A model sensitivity test reveals that this upward transport of the H<sub>2</sub>O plume is somewhat
- 848 dependent on the phase of the QBO at the time of the eruption. With the model QBO in a
- 849 westerly phase in January 2022 ("W" in Figure A6c), the accompanying circulation causes more
- rapid upward transport of the plume during the first half of 2022 compared to the easterly phase
- in Figure 6a-b. This upward transport during the westerly phase slows in late 2022 and into 2023
- as the QBO shifts to an easterly phase ("E" in Figure A6c). This simulation suggests that
- different phases of the QBO at the time of the eruption can cause a  $H_2O$  variation of  $\pm 1-3$  ppm in
- the equatorial mid-upper stratosphere, at least during the first  $\sim 1\frac{1}{2}$  years after the eruption
- 855 (Figure A6d). This dependence gradually fades after 2023 (not shown) as the excess  $H_2O$  decays
- and mixes throughout the global stratosphere.
- 857
- 858 A.4.3 Ozone
- 859

860 MLS observations show that ozone in the mid-stratosphere decreased significantly in the SH 861 subtropics and midlatitudes starting in early winter 2022. Mixing ratios decreased by up to 0.4-862 0.5 ppm (~10-12%) at 30-40 hPa in August 2022 (Figure A7a). Wang et al. (2022) suggest that 863 this anomalous low ozone was mainly due to circulation impacts (i.e., slowing of the BDC) 864 rather than chemical effects due to the HT SO<sub>2</sub> and H<sub>2</sub>O injection. GSFC2D model simulations 865 with the QBO and HT H<sub>2</sub>O anomaly included qualitatively reproduce many of the positive and 866 negative ozone anomalies seen in the MLS data averaged over 15°S-40°S (Figure A7b). 867 However, the anomaly magnitudes are generally underestimated in the model, and some of this 868 bias may be due to not including the HT sulfur injection in the simulation. Most of the model 869 anomalies are driven by circulation effects associated with the QBO (Figure A7c). However, the 870 impact of the HT  $H_2O$  anomaly in isolation is quite small, with ozone decreases of at most 0.03

871	ppm (-0.5%) at 30-40 hPa (Figure A7d). This result is generally consistent with Wang et al.
872	(2022) as most of the model ozone decrease at 15°S-40°S in winter 2022 is due to circulation
873	effects.
874 875	
876	Acknowledgments. The authors thank three anonymous reviewers for very helpful comments
877	and suggestions. This work was supported in part by the NASA Headquarters Atmospheric
878	Composition Modeling and Analysis Program.
879	
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884	Data Availability Statement. Total ozone satellite observations used in this manuscript
885	(Newman and Lait, 2023) are available on the NASA ozone watch website:
886	https://ozonewatch.gsfc.nasa.gov/.
887	The MERRA-2 reanalysis data products (GMAO, 2015) are available from the NASA Goddard
888	Earth Sciences Data and Information Services Center (GES DISC):
889	https://disc.gsfc.nasa.gov/datasets?project=MERRA-2.
890	The Aura/MLS data products are available from the NASA Goddard Earth Sciences Data and
891	Information Services Center (GES DISC) for H <sub>2</sub> O version 4 (Lambert et al., 2015) at:
892	https://doi.org/10.5067/Aura/MLS/DATA2009; H2O version 5 (Lambert et al., 2020) at:
893	https://doi.org/10.5067/Aura/MLS/DATA2508; and ozone version 5 (Schwartz et al., 2020) at:
894	https://doi.org/10.5067/Aura/MLS/DATA2516.
895	GSFC2D model description, configuration, input parameters and forcing datasets, and associated
896	references are provided in section 2 of the main text and Appendix A. GSFC 2D model output
897	used in the generation of the figures of this paper are available at
898	https://portal.nccs.nasa.gov/datashare/trop-str/pub/HT_H2O
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901	
902	
903	
904	
905	References
906	
907	Andrews, D. G., Holton, J. R., and Leovy, C. B., (1987). Middle atmosphere dynamics. 1st
908	Edition, 489, ISBN: 9780080211672. London. Academic Press
909	
910	Angell, J.K. (1993). Comparisons of stratospheric warming following Agung, El Chichón, and
911	Pinatubo volcanic eruptions, Geophysical Research Letters, 20, 715-718.
912	
913	Bacmeister, J. T., M. R. Schoeberl, M. E. Summers, J. E. Rosenfield, and X. Zhu (1995).
914	Descent of long-lived trace gases in the winter polar vortex, Journal of Geophysical Research -
915	Atmospheres, 100, 11,669-11,684.
916	
917	Baldwin, M.P., et al. (2001). The quasi-biennial oscillation, <i>Reviews of Geophysics</i> , <b>39</b> , 179-229.
918 919	Brasseur, G. and C. Granier (1992). Mount Pinatubo aerosols, chlorofluorocarbons, and ozone
920	depletion. <i>Science</i> . <b>257</b> , 1239-1242.
921	
922	Brasseur, G.P. and S. Solomon (2005). Aeronomy of the Middle Atmosphere, Springer
923	Netherlands, <u>https://doi.org/10.1007/1-4020-3824-0</u> .
924	
925	Burkholder, J. B, S. P. Sander, J. Abbatt, J. R. Barker, C. Cappa, J.D. Crounse, T.S. Dibble, R. E.
926	Huie, C. E. Kolb, M. J. Kurylo, V. L. Orkin, C.J. Percival, D. M. Wilmouth, and P. H. Wine
927	(2019). Chemical Kinetics and Photochemical Data for Use in Atmospheric Studies, Evaluation
928	No. 19, JPL Publication 19-5, Jet Propulsion Laboratory, Pasadena,
929	(http://jpldataeval.jpl.nasa.gov).
930	

- 931 Carr, J.L., Á. Horváth, D.L. Wu, and M.D. Friberg (2022). Stereo plume height and motion
- 932 retrievals for the record-setting Hunga Tonga-Hunga Ha'apai eruption of 15 January 2022.
- 933 Geophysical Research Letters, 49, e2022GL098131. https://doi.org/10.1029/2022GL098131 934
- 935 Clough, S. A., M. W. Shephard, E. J. Mlawer, J. S. Delamere, M. J. Iacono, K. Cady-Pereira, S.
- 936 Boukabara, and P. D. Brown (2005). Atmospheric radiative transfer modeling: A summary of the
- 937 AER codes, Journal of Quantitative Spectroscopy and Radiative Transfer, 91, 233–244.
- 938
- 939 Cohen, R.C., et al. (1994). Are models of catalytic removal of O<sub>3</sub> by HOx accurate? Constraints
- 940 from in situ measurements of the OH to  $HO_2$  ratio, Geophysical Research Letters, 21 (23), 2539-
- 941 2542.
- 942
- 943 Considine, D. B., A. R. Douglass, and C. H. Jackman (1994). Effects of a polar stratospheric
- 944 cloud parameterization on ozone depletion due to stratospheric aircraft in a two-dimensional 945 model, Journal of Geophysical Research – Atmospheres, 99, 18,879-18,894, 1994.
- 946
- 947 Coy, L., K. Wargan, A.M. Molod, W.R. McCarty, and S. Pawson (2016). Structure and
- 948 dynamics of the quasi-biennial oscillation in MERRA-2, Journal of Climate, 29, 5339-5354, 949 doi:10.1175/JCLI-D-15-0809.1
- 950
- 951 Coy, L., P.A. Newman, K. Wargan, G. Partyka, S.E. Strahan, and S. Pawson (2022).
- 952 Stratospheric circulation changes associated with the Hunga Tonga-Hunga Ha'apai eruption,
- 953 Geophysical Research Letters, 49, e2022GL100982. https://doi.org/10.1029/2022GL100982 954
- 955 Dunkerton, T.J. (1997). The role of gravity waves in the quasi-biennial oscillation, Journal of 956 *Geophysical Research – Atmospheres*, **102**, 26053-26076.
- 957
- 958 Dvortsov, V.L., and S. Solomon (2001). Response of the stratospheric temperatures and ozone to
- 959 past and future increases in stratospheric humidity, Journal of Geophysical Research –
- 960 Atmospheres, 106, 7505-7514.
- 961

962	Fleming, E.L., C.H. Jackman, R.S. Stolarski, and A.R. Douglas (2011). A model study of the
963	impact of source gas changes on the stratosphere for 1850-2100, Atmospheric Chemistry and
964	Physics, 11 (16), 8515-8541, doi:10.5194/acp-11-8515-2011.
965	
966	Fleming, E. L., P. A. Newman, Q. Liang, and J. S. Daniel (2020). The impact of continuing
967	CFC-11 emissions on stratospheric ozone. Journal of Geophysical Research: Atmospheres, 125,
968	e2019JD031849. https://doi.org/10.1029/2019jd031849
969	
970	Forster, P.M. de F., and K.P. Shine (1999). Stratospheric water vapour changes as a possible
971	contributor to observed stratospheric cooling, Geophysical Research Letters, 26, 3309-3312.
972	
973	Forster, P.M. de F., and K.P. Shine (2002). Assessing the climate impact of trends in
974	stratospheric water vapor, Geophysical Research Letters, 29 (6), 1086, 10.1029/2001GL013909.
975	
976	Garcia, R.R., D.R. Marsh, D.E. Kinnison, B.A. Boville, and F. Sassi (2007). Simulation of
977	secular trends in the middle atmosphere, 1950–2003, Journal of Geophysical Research:
978	Atmospheres, 112, D09301, doi:10.1029/2006JD007485.
979	
980	Gelaro, R., W. McCarty, M.J Suárez, R. Todling, A. Molod, L. Takacs, et al. (2017). The
981	Modern-Era retrospective analysis for Research and Applications, version 2 (MERRA-2).
982	Journal of Climate, 30(14), 5419-5454. https://doi.org/10.1175/jcli-d-16-0758.1
983	
984	Geller, M. A., et al. (2016). Modeling the QBO—Improvements resulting from higher-model
985	vertical resolution, Journal of Advance in Modeling Earth Systems, 8,
986	doi:10.1002/2016MS000699
987	
988	Gleason, J.F., P.K. Bhartia, J.R. Herman, R. McPeters, P. Newman, R.S. Stolarski, et al. (1993).
989	Record low global ozone in 1992, Science, 260 (5107), 523-526,
990	doi.org/10.1126/science.260.5107.523
991	

- 992 Global Modeling and Assimilation Office (GMAO) (2015). MERRA-2 inst3\_3d\_asm\_Np: 3d,3-
- 993 Hourly, Instantaneous, Pressure-Level, Assimilation, Assimilated Meteorological Fields V5.12.4
- 994 [Dataset], Greenbelt, MD, USA, Goddard Earth Sciences Data and Information Services Center
- 995 (GES DISC), Accessed: April 2023, https://doi.org/<u>10.5067/QBZ6MG944HW0</u>
- 996
- Gray, L.J., and J.A. Pyle (1989). A two-dimensional model of the quasi-biennial cycle of ozone, *Journal of the Atmospheric Sciences*, 46, 203-220.
- 999
- 1000 Hall, T.M. and D.W. Waugh, (1998). Influence of nonlocal chemistry on tracer distributions:
- 1001 inferring the mean age of air from SF<sub>6</sub>, *Journal of Geophysical Research Atmospheres*, **103**,
- 1002 13327–13336.
- 1003 Hall, T.M, D.W. Waugh, K. Boering, and R.A.Plumb (1999). Evaluation of transport in
- stratospheric models, *Journal of Geophysical Research Atmospheres*, **104**, 18815–18839.
  1005
- Hanson, D.R., A.R. Ravishankara, and S. Solomon (1994). Heterogeneous reactions in sulfuric
  acid aerosols: A framework for model calculations, *Journal of Geophysical Research* -
- 1008 *Atmospheres*, **99**, 3615-3629.
- 1009
- 1010 Hervig, M. E., U. Berger, and D. E. Siskind (2016). Decadal variability in PMCs and
- 1011 implications for changing temperature and water vapor in the upper mesosphere, *Journal of*
- 1012 *Geophysical Research Atmospheres*, **121**, 2383–2392, doi:10.1002/2015JD024439.
- 1013
- 1014 Hofmann, D.J., and S. Solomon (1989). Ozone destruction through heterogeneous chemistry
- 1015 following the eruption of El Chichón, *Journal of Geophysical Research*, 94, 5029-5041.
- 1016
- Holton, J.R., and X. Zhu (1984). A further study of gravity wave induced drag and diffusion in
  the mesosphere, *Journal of the Atmospheric Sciences*, 41, 2653-2662.
- 1019
- 1020 Hurwitz, M.M., E.L. Fleming, P.A. Newman, F. Li, E. Mlawer, K. Cady-Pereira, and R. Bailey
- 1021 (2015). Ozone depletion by hydrofluorocarbons, Geophysical Research Letters, 42(20), 8686-
- 1022 8692. doi:10.1002/2015GL065856.

1024	Jucks, K.W., et al. (1998). Observations of OH, HO <sub>2</sub> , H <sub>2</sub> O, and O <sub>3</sub> in the upper stratosphere:
1025	implications for HOx photochemistry, Geophysical Research Letters, 25 (21), 3935-3938.
1026	
1027	Khaykin, S., A. Podglajen, F. Ploeger, JU. Grooβ, F. Tence, S. Bekki, et al. (2022). Global
1028	perturbation of stratospheric water and aerosol burden by Hunga eruption. Communications
1029	Earth & Environment, 3(1), 316. https://doi.org/10.1038/s43247-022-00652-x
1030	
1031	Kiehl, J.T., B.A. Boville, and B.P. Briegleb (1988). Response of a general circulation model to a
1032	prescribed Antarctic ozone hole. Nature, 332, 501-504. https://doi.org/10.1038/332501a0
1033	
1034	Kovilakam, M., L.W. Thomason, N. Ernest, L. Rieger, A. Bourassa, and L. Millán (2020). The
1035	global space-based stratospheric aerosol climatology (version 2.0): 1979-2018, Earth System
1036	Science Data, 12, 2607-2634, https://doi.org/10.5194/essd-12-2607-2020
1037	
1038	Labitzke, K., and M.P. McCormick (1992). Stratospheric temperature increases due to Pinatubo
1039	aerosols, Geophysical Research Letters, 19, 207-210.
1040	
1041	Lambert, A., Read, W. and Livesey, N. (2015), MLS/Aura Level 2 Water Vapor (H2O) Mixing
1042	Ratio V004 [Dataset], Greenbelt, MD, USA, Goddard Earth Sciences Data and Information
1043	Services Center (GES DISC), Accessed: September 2023,
1044	https://doi.org/10.5067/Aura/MLS/DATA2009
1045	
1046	Lambert, A., Read, W. and Livesey, N. (2020), MLS/Aura Level 2 Water Vapor (H2O) Mixing
1047	Ratio V005 [Dataset], Greenbelt, MD, USA, Goddard Earth Sciences Data and Information
1048	Services Center (GES DISC), Accessed: September 2023,
1049	https://doi.org/10.5067/Aura/MLS/DATA2508
1050	
1051	Legras, B., C. Duchamp, P. Sellitto, A. Podglajen, E. Carboni, R. Siddans, et al. (2022). The
1052	evolution and dynamics of the Hunga Tonga - Hunga Ha' apai sulfate aerosol plume in the

1053 stratosphere. *Atmospheric Chemistry and Physics*, 22(22), 14957 – 14970.

1054 <u>http://doi.org/10.5194/acp-22-14957-2022</u>

1055

- Lindzen, R.S. (1981). Turbulence and stress owing to gravity wave and tidal breakdown, *Journal of Geophysical Research*, 86, 9707-9714.
- 1058 Lubken, F.-J., U. Berger, and G. Baumgarten (2018). On the anthropogenic impact on long-term
- 1059 evolution of noctilucent clouds. *Geophysical Research Letters*, 45, 6681–6689.
- 1060 https://doi.org/10.1029/2018GL077719
- 1061
- 1062 Mahlman, J.D., L.J. Umscheid, and J.P. Pinto (1994). Transport, radiative, and dynamical effects
- 1063 of the Antarctic ozone hole: A GFDL "SKYHI" model experiment. Journal of the Atmospheric
- 1064 Sciences, 51, 489–508. https://doi.org/10.1175/1520-0469(1994)051<0489:tradeo>2.0.co;2
- 1065
- 1066 Manney, G. L., M.L. Santee, A. Lambert, L.F. Millán, K. Minschwaner, F. Werner, et al. (2023).
- 1067 Siege in the southern stratosphere: Hunga Tonga-Hunga Ha'apai water vapor excluded from the
- 1068 2022 Antarctic polar vortex. *Geophysical Research Letters*, 50, e2023GL103855. https://doi.
- 1069 org/10.1029/2023GL103855
- 1070

1071 McCartney, E.J., *Optics of the Atmosphere*, John Wiley, New York, 1976.

- 1072
- McFarlane, N.A. (1987), The effect of orographically excited gravity wave drag on the general
  circulation of the lower stratosphere and troposphere, *Journal of the Atmospheric Sciences*, 44,
  1775–1800.
- 1076
- 1077 Meinshausen, M., Z.R.J. Nicholls, J. Lewis, M.J. Gidden, E. Vogel, M. Freund, U. Beyerle, C.
- 1078 Gessner, A. Nauels, N. Bauer, J.G. Canadell, J.S. Daniel, A. John, P.B. Krummel, G. Luderer, N.
- 1079 Meinshausen, S.A. Montzka, P.J. Rayner, S. Reimann, S.J. Smith, M. van den Berg, G.J.M.
- 1080 Velders, M.K. Vollmer, and R.H.J. Wang (2020). The shared socio-economic pathway (SSP)
- 1081 greenhouse gas concentrations and their extensions to 2500. Geoscientific Model Development,
- 1082 *13*(8), 3571-3605. doi:10.5194/gmd-13-3571-2020.
- 1083

1084	Millán, L., Santee, M. L., Lambert, A., Livesey, N. J., Werner, F., Schwartz, M. J., et al. (2022).
1085	Hunga Tonga-Hunga Ha'apai hydration of the stratosphere. Geophysical Research Letters, 49,
1086	e2022GL099381. https://doi.org/10.1029/2022GL09938
1087	
1088	Mlawer, E. J., S. J. Taubman, P. D. Brown, M. J. Iacono, and S. A. Clough (1997), RRTM: A
1089	validated correlated-k model for the longwave, Journal of Geophysical Research - Atmospheres,
1090	102, 16,663–16,682, doi:10.1029/97JD00237.
1091	
1092	Nevison, C.D., S. Solomon, and R.S. Gao (1999). Buffering interactions in the modeled response
1093	of stratospheric $O_3$ to increased $NO_X$ and $HO_X$ , Journal of Geophysical Research - Atmospheres,
1094	104, 3741-3754.
1095	
1096	Newman, P.A., and L.R. Lait (2023). NASA Ozone Watch: Images, data, and information for
1097	atmospheric ozone [Dataset]. National Aeronautics and Space Administration Goddard Space
1098	Flight Center. https://ozonewatch.gsfc.nasa.gov/.
1099	
1100	Plumb, R.A., and R.C. Bell (1982). A model of the quasi-biennial oscillation on an equatorial
1101	beta-plane, Quarterly Journal of the Royal Meteorological Society, 108, 335-352.
1102	
1103	Pollack, J., and T. Ackerman (1983). Possible effects of the El Chichón cloud on the radiation
1104	budget of the northern tropics, Geophysical Research Letters, 10, 1057-1060.
1105 1106	Portmann, R.W., S. Solomon, R.R. Garcia, L.W. Thomason, L.R. Poole, and M.P. McCormick
1107	(1996). Role of aerosol variations in anthropogenic ozone depletion in the polar regions, Journal
1108	of Geophysical Research - Atmospheres, 101, 22991-23006.
1109	
1110	Randel, W.J., F. Wu, J.M. Russel III, J. Waters, and L. Froidevaux (1995). Ozone and
1111	temperature changes in the stratosphere following the eruption of Mt. Pinatubo, Journal of
1112	Geophysical Research - Atmospheres, 100, 16753-16764.
1113	

1114	Randel, W. J., F. Wu, J.M. Russel III, A. Roche, and J. Waters (1998). Seasonal cycles and QBO
1115	variations in stratospheric CH4 and H2O observed in UARS HALOE data, Journal of the
1116	Atmospheric Sciences, 55(2), 163–185. https://doi.org/10.1175/1520-0469(1999)056<0457:
1117	GQCDFU>2.0.CO;2
1118	
1119	Randel, W.J., and F. Wu (1999). Cooling of the Arctic and Antarctic polar stratospheres due to
1120	ozone depletion. Journal of Climate, 12, 1467-1479. https://doi.org/10.1175/1520-
1121	0442(1999)012<1467:cotaaa>2.0.co;2
1122	
1123	Randel, W. J., Wu, F., Gettelman, A., Russell III, J. M., Zawodny, J. M., and Oltmans, S. J.
1124	(2001). Seasonal variation of water vapor in the lower stratosphere observed in halogen
1125	occultation experiment data, Journal of Geophysical Research - Atmospheres, 106, 14313-
1126	14325.
1127	
1128	Randeniya, L., P.F. Vohrolik, and I.C. Plumb (2002). Stratospheric ozone depletion at northern
1129	mid latitudes in the 21st century: The importance of future concentrations of greenhouse gases
1130	nitrous oxide and methane, Geophysical Research Letters, 29 (4), doi:10.1029/2001GL014295.
1131	
1132	Ray, E.A., F.L. Moore, J.W. Elkins, G.S. Dutton, D.W. Fahey, H. Vomel, S.J. Oltmans, and K.H.
1133	Rosenlof (1999). Transport into the Northern Hemisphere lowermost stratosphere revealed by
1134	insitu tracer measurements, Journal of Geophysical Research - Atmospheres, 104, 26565–26580.
1135	
1136	Rosenfield, J. E., D. B. Considine, P. E. Meade, J. T. Bacmeister, C. H. Jackman, and M. R.
1137	Schoeberl (1997). Stratospheric effects of Mount Pinatubo aerosol studied with a coupled two-
1138	dimensional model, Journal of Geophysical Research - Atmospheres, 102, 3649-3670.
1139	
1140	Rosenfield, J. E., and A. R. Douglass (1998). Doubled CO <sub>2</sub> effects on NOy in a coupled 2D
1141	model, Geophysical Research Letters, 25 (23), 4381-4384.

1143	Rosenfield, J. E., A. R. Douglass, and D. B. Considine (2002). The impact of increasing carbon
1144	dioxide on ozone recovery, Journal of Geophysical Research - Atmospheres, 107, 4049,
1145	doi:10.1029/2001JD000824.
1146	Schoeberl, M., Y. Wang, R. Ueyama, G. Taha, E. Jensen, and W. Yu (2022). Analysis and
1147	impact of the Hunga Tonga-Hunga Ha'apai stratospheric water vapor plume. Geophysical
1148	Research Letters, 49(20), e2022GL100248. https://doi.org/10.1029/2022GL100248
1149	
1150	Schoeberl, M., Y. Wang, R. Ueyama, G. Taha, and W. Yu (2023). The cross equatorial transport
1151	of the Hunga Tonga-Hunga Ha'apai eruption plume. Geophysical Research Letters, 50,
1152	e2022GL102443. https://doi.org/10.1029/2022GL102443
1153	
1154	Schwartz, M., Froidevaux, L., Livesey, N. and Read, W. (2020). MLS/Aura Level 2 Ozone (O3)
1155	Mixing Ratio V005 [Dataset], Greenbelt, MD, USA, Goddard Earth Sciences Data and
1156	Information Services Center (GES DISC), Accessed: September 2023,
1157	https://doi.org/10.5067/Aura/MLS/DATA2516
1158	
1159	Silletto, P., A. Podglagen, R. Belhadji, M. Boichu, E. Carboni, J. Cuesta, et al. (2022). The
1160	unexpected radiative impact of the Hunga Tonga eruption of January 15, 2022. Communications
1161	Earth & Environment, 3(1), 288. https://doi.org/10.1038/s43247-022-00618-z
1162	
1163	Solomon, S., R.W. Portmann, R.R. Garcia, L.W. Thomason, L.R. Poole, and M.P. McCormick
1164	(1996). The role of aerosol trends and variability in anthropogenic ozone depletion at northern
1165	midlatitudes, Journal of Geophysical Research - Atmospheres, 101, 6713-6727.
1166	
1167	Solomon, S. (1999). Stratospheric ozone depletion: A review of concepts and history, Reviews of
1168	Geophysics, 37, 275–316, doi:10.1029/1999RG900008.
1169	
1170	Solomon S., K.H. Rosenlof, R.W. Portmann, J.S. Daniel, S.M. Davis, T.J. Sanford, and GK.
1171	Plattner (2010). Contributions of stratospheric water vapor to decadal changes in the rate of
1172	global warming. Science, 327, 1219–1223. https://doi.org/10.1126/science.1182488

- 1174 Solomon, S., J. Haskins, D.J. Ivy, and F. Min (2014). Fundamental differences between Arctic
- and Antarctic ozone depletion, *Proceedings of the National Academy of Sciences*, 111(17),
- 1176 6220–6225. https://doi.org/10.1073/pnas.1319307111
- 1177
- 1178 Solomon, S., D. Kinnison, J. Bandoro, and R. Garcia (2015). Simulation of polar ozone
- depletion: An update, Journal of Geophysical Research Atmospheres, 120, 7958–7974,
- 1180 doi:10.1002/2015JD023365.
- 1181
- 1182 Stolarski, R.S., A.R. Douglass, M. Gupta, P.A. Newman, S. Pawson, M.R. Schoeberl, and J.E.
- 1183 Nielsen (2006). An ozone increase in the Antarctic summer stratosphere: A dynamical response
- to the ozone hole. *Geophysical Research Letters*, *33*, L21805.
- 1185 <u>https://doi.org/10.1029/2006gl026820</u>
- 1186
- 1187 Taha, G., R. Loughman, P. Colarco, T. Zhu, L. Thomason, and G. Jaross (2022). Tracking the
- 1188 2022 Hunga Tonga-Hunga Ha'apai aerosol cloud in the upper and middle stratosphere using
- space-based observations, *Geophysical Research Letters*, 49(19), e2022GL100091. https://doi.
- 1190 org/10.1029/2022GL100091
- 1191
- 1192 Thölix, L., A. Karpechko, L. Backman, and R. Kivi (2018). Linking uncertainty in simulated
- 1193 Arctic ozone loss to uncertainties in modelled tropical stratospheric water vapour, *Atmospheric*
- 1194 *Chemistry and Physics, 18,* 15047-15067, <u>http://doi.org/10.5194/acp-18-15047-2018</u>
- 1195
- Toon, O.B., P. Hamill, R.P. Turco, and J. Pinto (1986). Condensation of HNO<sub>3</sub> and HCl in the
  winter polar stratosphere, *Geophysical Research Letters*, *13*, 1284-1287.
- 1198
- 1199 Trepte, C. R., and M.H. Hitchman (1992). Tropical stratospheric circulation deduced from
- 1200 satellite aerosol data. *Nature*, *355*(6361), 626–628. https://doi.org/10.1038/355626a0
- 1201
- 1202 Vogel, B., T. Feck, and J.-U. Grooβ (2011). Impact of stratospheric water vapor enhancements

- 1203 caused by CH<sub>4</sub> and H<sub>2</sub>O increase on polar ozone loss, Journal of Geophysical Research -
- 1204 Atmospheres, 116, D05301, doi:10.1029/2010JD014234.
- 1205
- 1206 Vomel, H., S. Evan, and M. Tully (2022). Water vapor injection into the stratosphere by Hunga
- 1207 Tonga-Hunga Ha'apai. Science, 377(6613), 1444–1447. <u>https://doi.org/10.1126/science.abq2299</u>
  1208
- 1209 Wallace, J.M. (1973). General circulation of the tropical lower stratosphere, *Reviews of*
- 1210 *Geophysics*, 11(2), 191-222.
- 1211
- 1212 Wang, X., W. Randel, Y. Zhu, et al. (2022). Stratospheric climate anomalies and ozone loss
- 1213 caused by the Hunga Tonga volcanic eruption. ESS Open Archive,
- 1214 doi: 10.1002/essoar.10512922.1, <u>https://essopenarchive.org/doi/full/10.1002/essoar.10512922.1</u>
- 1215
- Witze, A. (2022). Why the Tongan eruption will go down in the history of volcanology. *Nature*,
  376–378. <u>https://doi.org/10.1038/d41586-022-00394-y</u>
- 1218
- 1219 World Meteorological Organization (WMO, 2022). Scientific Assessment of Ozone Depletion:

1220 2022, GAW Report No. 278, 509 pp.; WMO: Geneva, 2022.

- 1221
- 1222 Zhu, Y., C.G. Bardeen, S. Tilmes, et al. (2022). Perturbations in stratospheric aerosol evolution
- due to the water-rich plume of the 2022 Hunga-Tonga eruption. *Communications Earth and*
- 1224 Environment 3, 248 (2022). https://doi.org/10.1038/s43247-022-
- 1225 00580-w
- 1226
- 1227
- 1228
- 1229
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# 1237 Figure captions

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1239 Figure 1. Top panels: water vapor anomaly derived from MLS v4 observations for the months

indicated. Contours are 0.1, 0.5, 1, 2, 4, 6, and 8 ppm. *Bottom panels*: model temperature

1241 response, taken as the difference between simulations with and without the water vapor

anomaly. Negative contours are -3, -2, -1, -0.5, and -0.2 K, and positive contour intervals are in

- 1243 0.1K increments. The thick black solid line indicates the tropopause.
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Figure 2. *Top panels*: monthly mean de-seasonalized temperature (color shading) and residual circulation (white streamlines) from the Modern-Era Retrospective Analysis for Research and Applications, version 2 (MERRA-2) for May (left) and August (right) 2022. *Bottom panels*: deseasonalized temperature normalized by the standard deviation (color shading). See text for details. The letters show the locations of the time series in Figure 3.

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1251 Figure 3. Daily de-seasonalized temperature for 2010 through 2022 from the MERRA-2 1252 reanalysis (black lines) at 20°S, 20 hPa (top) and 40°S, 27 hPa (bottom), the locations of "A" and 1253 "B", respectively, shown in Figures 2 and 4. Two model simulations are included: 1) with an 1254 interactive QBO and no water vapor anomaly (blue line); and 2) with both the QBO and water 1255 vapor anomaly (red line). The vertical magenta lines denote the date of the eruption (15 1256 January 2022). The model is free running and does not correspond to a particular year, except 1257 following the eruption when the model zonal wind QBO phase is similar to the Singapore 1258 radiosonde observations and MERRA-2. The right-hand axes show the de-seasonalized 1259 temperature in terms of the number of standard deviations.

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Figure 4. May 2022 average de-seasonalized temperature (color shading) and de-seasonalized
residual circulation (red streamlines). Shown are (a) MERRA-2 reanalysis (repeated from Figure

1263	2a), and (b)-(d) model simulations that include an interactive QBO: (b) with the water vapor
1264	anomaly; (c) no water vapor anomaly; and (d) the difference, (b) minus (c). The model with
1265	both the QBO and water vapor anomaly (b) best matches MERRA-2 (a). The letter "A" shows
1266	the location of the time series in Figure 3a.

1268 **Figure 5.** Time-latitude cross-sections of the model daily water vapor and temperature

anomalies for the altitudes indicated. The vertical dotted line in panel (a) denotes the change

1270 from the MLS v4 H<sub>2</sub>O anomaly to model output on 1 January 2023. Contour intervals are: (a)

1271 .02, .05, .1, .2, .5, 1, 2, 5, 10, and 15 ppm; (b)-(c): ±1K and includes the ±0.4K contours.

1272

Figure 6. Time-altitude cross-sections of the model daily globally averaged (a) water vapor, (b)
temperature, and (c) ozone anomalies. The vertical dotted line in the top panel denotes the
change from the MLS v4 water vapor anomaly to model output on 1 January 2023. Contour
intervals are: (a) .05, .1, .2, .5, 1, and 2 ppm; (b): ±0.1, -0.5, and -1K; (c): ±0.5, -1, -2, -5, and
-10%.

1278

Figure 7. Daily total global water vapor burden anomaly (teragrams, Tg, black line), and a fitted
decay of the simulated burden from 1 July 2023 assuming a constant first order loss with a
global lifetime of 2.5 years (red dashed-dotted line). The vertical dotted line denotes the
change from the MLS v4 anomaly to model output on 1 January 2023.

1283

Figure 8. Model anomalies of (a) H<sub>2</sub>O, (b) OH, (c) temperature, (d) NOx, and (e) ozone, averaged
over a two-year (2023-2024) period. Contour intervals are: (a) +5%; (b): +2%; (c): -1, -0.5, 0.05,
0.1K; (d) and (e): -5, -2, +1%. The thick black solid line indicates the tropopause.

1287

1288 Figure 9. Model global mean vertical profiles of anomalies averaged over a two-year (2023-

1289 2024) period. Shown are: (a) concentrations of the constituents indicated; (b) tendency of odd

1290 oxygen (O+O<sub>3</sub>) due to the chemical loss cycles of the odd hydrogen (HOx), odd nitrogen (NOx),

1291 odd oxygen (Ox), and chlorine (ClOx) families, and the total chemical tendency.

1292

Figure 10. Time-altitude cross-sections of daily model (a) water vapor, (b) temperature, and (c)
ozone anomalies averaged over the Antarctic polar cap (65°S-90°S), and (d) daily MLS v5 ozone
through 30 November 2023 averaged over 65°S-80°S with the 2005-2021 average seasonal
cycle removed. The vertical dotted line in panel (a) denotes the change from the MLS v4 water
vapor anomaly to model output on 1 January 2023. Contours are: (a) 0.1, 0.2, 0.5, 1, and 2 ppm;
(b): intervals of ±0.5K and includes the ±0.1 and ±0.2K contours; (c) and (d): intervals of ±1
DU/km and includes the ±0.2 and ±0.5 DU/km contours.

1300

Figure 11. April-December 2023 daily model anomalies averaged over the Antarctic polar cap ( $65^{\circ}S-90^{\circ}S$ ) at 54 hPa (20 km) for (a) water vapor and ice PSCs, (b) nitrogen species (ppb, solid lines) and the combined rate of the heterogeneous reaction ClONO<sub>2</sub>+HCl $\rightarrow$ HNO<sub>3</sub>+Cl<sub>2</sub> on NAT and Ice PSCs and sulfate aerosols (1/day, dotted line), (c) odd oxygen loss (mainly ozone), (d) ozone, odd oxygen loss and transport, and (e) temperature and solar ozone heating. The righthand axis indicates the odd oxygen tendencies in panel (d), and the solar ozone heating rate in panel (e).

1308 1309

Figure 12. Model anomalous odd oxygen (O+O<sub>3</sub>) tendencies over the Antarctic polar cap (65°S90°S) at 28 hPa (25 km) for 2023. Shown are the daily chemical tendencies due to the odd
hydrogen (HOx, green), odd nitrogen (NOx, blue), chlorine (ClOx, red), and bromine (BrOx,
orange) families. Also shown is the tendency due to the transport of odd oxygen (black dash dot
line).

1315

Figure 13. Daily model total ozone anomalies over the 2022-2031 period. Contour intervals are
-5 DU and include the ±1 DU contours to show minor effects.

1318

Figure 14. Daily total ozone seasonal cycle for (a) 2023 Antarctic (63°S-90°S) and (b) July 2023 June 2024 for the Arctic (63°N-90°N). Shown are model simulations under different planetary
wave forcing conditions (colored lines) for the baseline (solid) and including the HT water vapor
anomaly (dashed). The gray shades depict satellite climatology (1991-2022, Newman and Lait,
2023).

Figure A1. Time series of equatorial stratospheric zonal mean zonal wind from the (a) MERRA-2
reanalysis for 1993-2005, and (b) model simulation. Contour intervals are ±10 m/sec, with the
westerlies in red and the easterlies in blue.

1328

Figure A2. Age of air at: (a) 20 km (~50 hPa) derived from ER-2 aircraft observations of SF<sub>6</sub> (blue asterisks) and CO<sub>2</sub> (red triangles), and (b)-(d) vertical profiles of the age of air derived from balloon measurements of SF<sub>6</sub> (blue asterisks, green plus signs) and CO<sub>2</sub> (red triangles) at the latitudes indicated (adapted from Hall et al., 1999). The observations are taken during the 1990s. Also shown is the model age of air annually averaged over the 1990s (black line). The age is taken relative to the tropical tropopause.

1335

Figure A3. Monthly mean water vapor (ppm) for June and October 2022 from MLS v5 (toppanels) and model simulation (bottom panels). The contour interval is 1 ppm.

1338

Figure A4. Time-latitude cross-sections of daily water vapor (ppm) at 28 hPa for January 2022 November 2023 from (a) MLS v5 and (b) model. The model is forced with the MLS-derived H<sub>2</sub>O
 anomaly prior to 1 March 2022 (vertical dotted line in panel (b)). The contour interval is 1 ppm.

Figure A5. Time-altitude cross-sections of daily water vapor (ppm) averaged over 65°S-80°S for
January 2022-November 2023 from (a) MLS v5 and (b) model. The contour interval is 1 ppm.

1346 Figure A6. Time-altitude cross-sections of daily water vapor (ppm) averaged over 10°S-10°S for 1347 January 2022-November 2023 from: (a) MLS v5, and model simulations with the zonal wind 1348 QBO in the same phase (b) and roughly 180° out of phase (c) with the Singapore radiosonde 1349 observations. Panel (d) shows the model difference, (c) minus (b). Maximum easterly ("E") and 1350 westerly ("W") zonal winds at 30 hPa are from the Singapore observations in panel (a) and the 1351 respective model simulations in panels (b) and (c). The model is forced with the MLS-derived 1352 H<sub>2</sub>O anomaly prior to 1 March 2022 (vertical dotted line in panels (b)-(d)). The contour interval 1353 is 1 ppm.

- 1355 **Figure A7.** Time-altitude cross-sections of daily de-seasonalized ozone (ppm) averaged over
- 1356 15°S-40°S. Shown are (a) MLS v5 data, and (b)-(d) model simulations that include an interactive
- 1357 QBO: (b) with the water vapor anomaly; (c) no water vapor anomaly; and (d) the difference, (b)
- 1358 minus (c). The contour interval is ±0.1 ppm. The vertical magenta lines denote the date of the
- 1359 eruption (15 January 2022).