

#### **Abstract.**

 The January 2022 eruption of the Hunga Tonga-Hunga Ha'apai underwater volcano injected a 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 temperature changes occur in the Southern hemisphere (SH) subtropics in April-May 2022, with warming of ~1K in the lower stratosphere and cooling of 3K in the mid-stratosphere. The radiative cooling combined with adiabatic cooling driven by the quasi-biennial oscillation 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 44 transported globally throughout the stratosphere and mesosphere. The excess  $H_2O$  increases the OH radical, causing a negative global ozone response (2-10%) in the upper stratosphere and mesosphere due to increased odd hydrogen-ozone loss, and a small positive ozone response (0.5- 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 and springtime halogen-ozone loss, enhancing the Antarctic ozone hole by 25-30 DU in 2023. 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<sub>2</sub>O is removed from the stratosphere with a 2.5-year e-folding time. Given the year-to-year variability of the stratosphere, the magnitudes of these ozone responses may be below the threshold of detectability in observations. Key Words: stratosphere, ozone depletion, Hunga Tonga Hunga Ha'apai eruption, water vapor, greenhouse gas 

#### **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

modest amount of SO<sup>2</sup> 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
- examine the dependence of the polar ozone response on background stratospheric conditions.
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- **2. Model Simulations**
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# **2.1 Model description**

 Simulations in this study are conducted with the National Aeronautics and Space Administration (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 Meteorological Organization ozone assessments, including WMO (2022). The model has been described and evaluated previously and has been shown to provide realistic simulations of ozone, temperature, and transport-sensitive tracers for a variety of stratospheric perturbations (Bacmeister et al., 1995; Jackman et al., 1996, 2016; Rosenfield et al., 1997, 2002; Fleming et al., 2011; 2020). The model has very small internal variability, so that responses to small perturbations can be easily detected. In Appendix A, we provide a description of recent updates and model components important to the present study, including simulation of the quasi-biennial oscillation (QBO). We also provide an evaluation and comparison of the model with observations.

## **2.2 Water vapor simulation**

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

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 12 161 km to the tropopause. This unperturbed H<sub>2</sub>O does not interact with other model components.
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 To simulate the HT water vapor perturbation in the model, we use MLS version 4 (v4) data as input. Previous analysis suggests that MLS v4 provides a more suitable data product to estimate the HT H2O anomaly compared to the most recent version 5 data (Millán et al., 2022). We determine the zonal mean MLS water vapor anomaly in the latitude-height domain for 15 January 2022 through 31 December 2022 as the difference for each day from the 2005-2021 average for a given month. The difference of the pre-eruption 1-14 January 2022 average from 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 MLS quality and convergence filters flagged some profiles during the first few weeks after the eruption, however, the water vapor enhancements were independent of the quality and convergence filtering after 8 February (Millán et al., 2022). 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<sub>2</sub>O tracer that interacts with the model chemistry, radiation, and 177 dynamics. This perturbed interactive  $H_2O$  is updated in this manner on 15 January through 31 December 2022. This simulates the evolving impacts of the water vapor anomaly through the end of 2022 using the MLS observations as input at each time step. Starting 1 January 2023, the evolution of the perturbed H2O is fully model-computed, with the MLS-based field on 31 December 2022 serving as the initial condition. We also ran a parallel baseline simulation without the MLS-derived HT water vapor anomaly. The stratospheric response to the HT H2O perturbation is taken as the difference between the perturbation and baseline simulations. 

**3. Results**

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

 The water vapor anomaly mixes and disperses throughout the global stratosphere during 2022, as seen in the MLS-derived observations for selected months (Figure 1, top panels). While the initial H2O injection at 20°S reached the upper stratosphere on 15 January (Carr et al., 2022), by April the anomalous water vapor was mainly confined to the mid-stratosphere in a latitude band from ~40°S to 25°N. By August and especially December, the upward tropical bulge and downward midlatitude bulge of the plume reflect transport by the Brewer-Dobson circulation (BDC) as noted previously in Schoeberl et al., 2022. The Southern hemisphere (SH) polar vortex 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 into the polar region following the vortex break-up in late November and December (Figure 1e).

 The corresponding model temperature response is significant and is highly correlated with the H2O anomaly (Figure 1, bottom panels). The increased stratospheric water vapor enhances the IR 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  $\sim$ 1K at 54 hPa. Warming of up to several tenths of a degree K occurs around the tropical tropopause. The temperature response spreads vertically and horizontally and decreases in magnitude throughout 2022, following the dispersal of the water vapor plume in latitude and altitude (Figure 1).

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

215 In this section we examine how the model temperature response to the HT  $H_2O$  anomaly

216 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.



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

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 [\(https://acd-](https://acd-ext.gsfc.nasa.gov/Data_services/met/qbo/qbo.html#singau)
- [ext.gsfc.nasa.gov/Data\\_services/met/qbo/qbo.html#singau\)](https://acd-ext.gsfc.nasa.gov/Data_services/met/qbo/qbo.html#singau). The corresponding model
- meridional QBO circulation and associated temperature anomalies are qualitatively consistent
- 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
- 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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 Including both the QBO and the water vapor anomaly brings the model into very good agreement with the MERRA-2 mid-stratosphere warm and cold anomalies in the tropics and SH low-mid latitudes (Figures 4a-b). The model also captures quite well the MERRA-2 extreme cold anomaly at 20°S, 20 hPa following the eruption through May 2022, as well as the return to near normal seasonal temperatures by the end of August 2022 (Figure 3a, red line). For the cold 268 temperature anomaly at  $20^{\circ}$ S, 20 hPa in May 2022 ( $\sim$ -5K), we estimate from the model that  $\sim$  60% (-3K) is caused by IR cooling of the excess H<sub>2</sub>O, and  $\sim$  40% (-2K) is caused by ascent of 270 the QBO-induced circulation. The model with both the QBO and  $H_2O$  anomaly included also qualitatively captures the warm and cold anomalies in MERRA-2 in the lower stratosphere below ~40 hPa at 30°S-30°N (Figure 4a-b). These model simulations confirm that the very cold mid-stratosphere anomaly in the SH subtropics in May 2022 is caused by the HT water vapor anomaly but is reinforced by the ascent and adiabatic cooling caused by the meridional circulation associated with the QBO being in an easterly phase in May 2022.

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

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
- 281 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.

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

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

 The excess H2O is transported to the SH mid-high latitudes in the months following the eruption, 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  $H_2O$  (starting 1 January 2023) maintains concentrations 301 of  $\sim$ 2.5 ppm throughout the SH high latitudes in January 2023, before slowly diminishing to  $\sim$ 2 ppm by March 2023. This decrease is due to the slow removal of the excess stratospheric water 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 latitudes (Figure 5a) with the establishment of the polar vortex and onset of PSC formation. The negative anomalous mixing ratios are due to the increased sedimentation of ice PSCs with the enhanced water vapor relative to the baseline. Positive anomalous mixing ratios return to the SH polar region in spring 2023 with the breakup of the vortex and in-mixing of mid-latitude air. 

310 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 ~1 ppm poleward of 60°N throughout March and April 2023.

- The model temperature response at 25 hPa (Figure 5b) is highly correlated with the H2O 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
- confined to the tropics and SH lower latitudes in 2022, with a small warming of 0.1-0.2K outside
- the polar regions (50°S-50°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
- mid-2029. Mostly small cooling of 0.1-0.2K occurs throughout the stratosphere after 2023,
- 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.
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 For 1-3 years following the eruption, the water vapor anomaly is slowly transported upwards by the BDC, with mixing ratios of 1-1.5 ppm reaching the mesosphere by late 2023 (Figure 6a). Here we show the global average since the anomalies generally have similar patterns across most latitude zones (the ozone response in Figure 6c will be discussed in section 3.4). Global 329 temperature changes again follow the H<sub>2</sub>O plume, with cooling of  $\sim$ 1K confined to the mid- stratosphere during 2022 and cooling of 1-1.5K in the mesosphere in mid-2023 through the end of 2024 (Figure 6b). Small global-mean warming of 0.1-0.15K occurs in the upper troposphere/lower stratosphere just after the eruption through late 2023. Starting in 2024, the global average temperature response is almost exclusively negative (cooling) throughout the middle atmosphere as the anomalies gradually diminish to be quite small by 2030. 336 The total global burden (mass) of the H<sub>2</sub>O anomaly is 150-160 Tg shortly after the eruption through mid-2023 and is projected to decrease thereafter (Figure 7). The excess stratospheric water vapor is slowly removed by sedimentation of PSCs within the Antarctic vortex, as well as return of stratospheric air to the troposphere by the BDC at mid-high latitudes of both hemispheres. The combination of these processes leads to an exponential decay of the anomaly, 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  $\sim$ 3% of its starting value in January 2022. 

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

 Ozone is affected by the HT water vapor anomaly globally throughout the middle atmosphere due to changes in photochemistry, both directly, and indirectly via changes in the temperature- dependent ozone loss cycles (e.g., Dvortsov and Solomon, 2001; Brasseur and Solomon, 2005). MLS observations show that ozone in the mid stratosphere decreased significantly in the SH 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 small in 2022. This is discussed further in section A.4.3 of Appendix A, along with comparisons to the MLS data.

 For globally averaged ozone, maximum projected changes occur in 2023-2024, ~1-2 years after the eruption (Figure 6c). During this time, the H2O anomaly is 20-25% above the background in the SH mid-stratosphere, and ~15% above the background globally throughout the mesosphere 359 (Figure 8a). One of the direct consequences of excess  $H_2O$  is the increase in odd hydrogen 360 species (HOx = H+OH+HO<sub>2</sub>+2\*H<sub>2</sub>O<sub>2</sub>), with the OH radical increasing by 5-10% throughout the 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 km (Figure 9b). The resulting projected global ozone loss is >0.5% at altitudes above ~40 km throughout 2023-2026 (Figure 6c) and increases with altitude to 5-10% above ~60 km from mid-2023 to mid-2025.

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,

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

370 (NOx=N+NO+NO<sub>2</sub>+NO<sub>3</sub>+2<sup>\*</sup>N<sub>2</sub>O<sub>5</sub>; Figure 9a, blue line) are mostly negative throughout the

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

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

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

 $377 \text{ N}_2\text{O}_5+\text{H}_2\text{O} \rightarrow 2*$  HNO3 on sulfates. Although the model stratospheric sulfate aerosol surface area is specified and does not interact with the H2O 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.

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 384 reaction N+O<sub>2</sub>  $\rightarrow$  NO+O at lower temperatures. The increased N increases the loss of NO<sub>y</sub> which 385 is controlled by the reaction N+NO $\rightarrow$ N<sub>2</sub>+O (Rosenfield and Douglass, 1998).

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

398 In the lower stratosphere below  $\sim$ 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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#### **3.5 Antarctic profile ozone and related chemical responses**

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

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

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

- temperatures in the polar vortex (Wang et al., 2022). For the model simulations presented in this
- study, the SH planetary wave forcing (section A.2) was reduced during winter-spring 2022 to
- mimic the strong and isolated SH polar vortex that persisted well into November. However, the
- 411 resulting additional ozone depletion due to the HT  $H_2O$  anomaly was significantly smaller than
- shown in the de-seasonalized MLS data in spring 2022 (Figure 10c-d). The model shows some
- qualitative consistency with the MLS ozone during the first half of 2023, with a negative
- anomaly at 10-20 km and a positive anomaly at 20-25 km.
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In late winter-spring 2023, the model simulates a deepened ozone hole, with a significant

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

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

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

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

impact of the excess H2O in the de-seasonalized MLS ozone data. Model biases, at least for the

- first 1-1½ years after the eruption, also may be due in part to not including the HT aerosol
- perturbation in the model.
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425 The processes that cause the ozone hole have been well established in past studies (e.g., Solomon, 1999; Solomon et al., 2014, Solomon et al., 2015). To examine in more detail how the excess H2O impacts the model ozone hole, we focus on lower stratospheric anomalies for April-December 2023 of several constituents relevant to the chemistry driving the enhanced ozone

- hole. By mid-winter 2023, the water vapor anomaly is present within the Antarctic vortex
- 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

- type II ice PSCs throughout the winter (Figures 11a-b). The increased sedimentation of NAT
- PSCs causes denitrification (e.g., Toon et al., 1986) starting in early June, with negative
- anomalous NOy persisting through winter and spring and negative anomalous NOx

concentrations in October-November (Figure 11b).

 The projected enhancement in PSCs and PSC surface area increases the heterogeneous conversion of chlorine and bromine from reservoir species (HCl, ClONO2, HOBr, BrONO2) to reactive forms that destroy ozone (Cl, ClO, Br, BrO). The anomalous heterogeneous chlorine activation on sulfate aerosols is also generally faster due to the lower temperatures and increased water vapor (e.g., Solomon, 1999; Burkholder et al., 2019), even though the model sulfate aerosol surface area is specified and does not interact with the H2O anomaly. The anomalous rate 444 of a key heterogeneous reaction,  $CIONO_2 + HCl \rightarrow HNO_3 + Cl_2$  on PSCs and sulfates, is shown in Figure 11b (purple dotted line). The anomaly maximizes in June following the anomalies in PSC surface area and enhancement of the reaction on sulfates. The secondary anomaly maximum 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 H2O anomalies are all quite small in the spring. Increased conversion of chlorine to reactive forms on sulfate aerosols under cold SH polar conditions was noted previously (e.g., Hanson et al., 1994).

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

 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.

 In the middle stratosphere (~40-10 hPa), positive model ozone anomalies in the Antarctic occur from December 2022-June 2023 (Figure 10c), mainly due to the reduced NOx catalytic loss cycle (i.e., increased odd oxygen tendency, Figure 12 blue line). A negative ozone anomaly driven by the increased halogen loss cycles occurs during August-September 2023. Relatively small ozone changes occur during late September-October as the positive transport tendency largely offsets the anomalous chemical ozone loss. Positive ozone anomalies re-emerge in the mid-stratosphere during December 2023 through June 2024 (Figure 10c), driven by a combination of reduced NOx loss and positive anomalous transport tendency. This anomalous polar transport occurs during the SH late spring-summer and is a dynamical response to the enhanced ozone hole. This feature has been discussed in previous modeling studies (Kiehl et al., 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, the additional ozone depletion leads to a projected delay in 1) the spring vortex breakup, and 2) the corresponding wave-forced drag on the zonal mean flow and acceleration of the BDC. As a result, there is anomalous descent in the Antarctic mid-upper stratosphere and positive odd oxygen transport tendency relative to the baseline in late November-December 2023 (Figure 12). The anomalous descent and associated adiabatic warming cause the positive temperature anomaly in the polar mid-stratosphere above ~40hPa during November-December 2023 (Figure 10b). This warm anomaly is a yearly recurring feature in the Antarctic summer mid-stratosphere throughout 2023-2030 (Figure 5b).

 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.

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

## 3.6.1 Baseline response



- 520 and 1-2 DU in the NH, with changes of less than  $\pm 1$  DU in the tropics throughout the post-
- eruption period.
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3.6.2 Sensitivity to stratospheric background conditions

 The projected ozone responses shown in Figures 13 are from a model simulation that uses the standard planetary wave forcing which gives climatologically averaged stratospheric conditions (section A.2). This simulation is generally consistent with the observed long term average seasonal cycle in polar total ozone in each hemisphere (Figure 14, orange vs. black solid lines). However, the polar regions are characterized by large interannual variability in winter and spring 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 for 1991-2022 (Newman and Lait, 2023).

 To examine the dependence of the ozone response on the background stratospheric conditions, we ran a series of experiments with the stratospheric wave driving varied to mimic the observed range in total ozone shown in Figure 14. Here we focus on the model year with the largest anomalous ozone loss in each polar region: 2023 for the SH and July 2023-June 2024 for the NH. Substantially increased planetary wave forcing gives warm polar stratospheric conditions and total ozone at the upper end of the range in the historical data record in both hemispheres (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 wave driving case, the water vapor anomaly simulation is depicted by the dashed lines in Figure 14.

545 In the SH, including the HT  $H_2O$  perturbation under the strong wave forcing (warm) conditions results in somewhat less projected ozone depletion compared to the standard wave forcing, with polar cap average additional depletion of 13 DU vs. 20 DU in 2023 (Figure 14a). The stronger wave driving, and warmer stratospheric conditions also promote a faster return to the baseline total ozone in December 2023 compared to the standard case. Under the weak wave forcing (cold) conditions, the baseline already has very low ozone concentrations in the lower 551 stratosphere, so that including the  $H_2O$  anomaly has less of an impact than with the standard wave forcing, with additional polar cap average depletion of 14 DU (Figure 14a, blue dashed line). The largest impact occurs under conditions slightly warmer than the standard case (not shown). Here, the larger ozone concentrations available in the baseline combined with substantial anomalous chlorine and bromine activation resulted in additional polar cap depletion of 23 DU, compared to 20 DU for the standard wave forcing case.

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

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

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

 the cold conditions of the weak wave driving case, the additional model ozone depletion in the 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 PSC formation in early winter (November-December 2023), the excess water vapor is 0.2-1 ppm in the Arctic lower stratosphere, substantially less than the 1-2 ppm in the early winter Antarctic (May-June 2023). Another important factor is the generally warmer Arctic temperatures that limit additional ozone depletion (Solomon et al., 2014). For example, in the weak wave driving (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 consistent with previous studies of the Arctic ozone response to stratospheric water vapor changes using 3-D chemical transport models driven by meteorological reanalysis (e.g., Vogel et al., 2011; Thölix et al., 2018).

 Figure 14 suggests that the total ozone impact of the HT H2O anomaly is generally smaller than the year-to-year variability characteristic of the stratospheric polar regions during winter and spring. The standard deviation of the observed SH October-November polar cap total ozone is ~35 DU during 1991-2022, significantly larger than the maximum model estimated response to the H2O anomaly of 23 DU. The difference is even larger in the Arctic spring, with a March- April observed standard deviation of ~26 DU compared to a maximum model anomaly response of 5 DU. Therefore, it is possible that the response to the HT water vapor injection may not be easily detectable above the background variability in observational total ozone data.

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## **4. Summary and Conclusions**

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

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;
- 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 1- 2 years following the eruption and estimate possible future responses over the next decade.

 The maximum radiatively-induced model temperature response occurs in March-June 2022, several months after the eruption, with a cooling of 2-3K in the SH mid-stratosphere, and ~1K warming in the lower stratosphere. This radiatively-driven warming is as much as several tenths of a degree K around the tropical tropopause, which may have important implications for the amount of water vapor entering the stratosphere. However, quantification of this effect is beyond the scope of the 2D model used in this study. We note that these impacts do not include sea surface temperature feedback, which may be important to fully quantify the response to the 602 radiative forcing of the HT  $H_2O$  anomaly.

 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 606 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.

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

 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 HNO3, 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 catalytic cycle dominates the chemical loss of odd oxygen.

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

 By mid-2023 and beyond, the excess H2O is slowly removed by return to the troposphere at mid- high 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 641 and ozone responses diminish slowly after 2024. By the end of 2031, the additional  $H_2O$  is 642 estimated to be  $\sim$ 3% of its initial value of  $\sim$ 150 Tg, with very small temperature and ozone responses.

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  $SO<sub>2</sub>(0.4-1)$  0.5 Tg) which likely increased the stratospheric sulfate aerosol layer (Legras et al., 2022; Taha et al., 2022; Zhu et al., 2022). This could impact ozone chemistry via increased heterogeneous conversion of odd nitrogen to HNO3, and increased chlorine activation at lower temperatures (~195K). Stratospheric temperatures can also be affected via changes in ozone and aerosol absorption of infrared radiation. These impacts can occur globally and in the polar regions as shown for previous volcanic eruptions (e.g., Brasseur and Granier, 1992; Randel et al., 1995; Solomon et al., 1996; Portmann et al., 1996). For the HT eruption, Wang et al. (2022) showed

 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 eruption, at least in the first year following the eruption. The model-projected ozone responses presented here are generally smaller than the natural variability of the stratosphere. The largest response is likely the enhancement of the Antarctic ozone hole. However, the additional model ozone loss is at most 23 DU (2023 polar cap average), which is smaller than the standard deviation of springtime polar total ozone 663 observations in the SH ( $\sim$ 35 DU). Therefore, it is possible that any ozone response to the HT water vapor injection may not be detectable above the background variability in observational data sets. **Appendix A: GSFC2D Model Description and Evaluation** In this appendix, we provide a description and evaluation of the GSFC2D model, focusing on recent updates and model components important to the present study. We also provide some model evaluation via comparisons with observations of age of air, H2O, and ozone. A.1 Model chemistry and radiation The model has full stratospheric chemistry, with a diurnal cycle computed for all constituents each day. Transport of all constituents follows the chemistry calculations at each time step within the diurnal cycle. This is updated from the previous scheme in which only the diurnal averages were transported at the end of the diurnal cycle. The resulting changes are most significant at mid-high latitudes during late autumn through early spring when the photochemical time scales are long, and the new methodology improves the simulations of polar ozone compared with 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.

 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 688  $CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O. The latest JPL-2019 recommendations (Burkholder et al., 2019) are used for$  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).

 A parameterization to simulate type Ia (solid nitric acid trihydrate, NAT) and type II (Ice) PSC formation follows Considine et al. (1994) and includes sedimentation of NAT and ice aerosols. Calculations of PSC occurrence frequency and surface area density use a combination of the zonal mean temperatures computed in the model and longitudinal temperature probability distributions (deviations from the zonal mean, T΄) obtained from the MERRA-2 reanalysis. Here, the MERRA-2 climatological T΄ distribution is based on daily data averaged over 2000-2020 and is added to the model-computed zonal mean temperatures at each time step. This hybrid methodology allows the PSCs to respond to both observational-based longitudinal variations and the evolving model zonal mean temperature. To convert the PSC concentration to surface area density, a lognormal particle size distribution is assumed (Considine et al., 1994). The uptake coefficients for heterogeneous reactions on PSCs are specified to be constant (Burkholder et al., ). We note that supercooled ternary solution (STS,  $H_2SO_4/HNO_3/H_2O$ ) type Ib PSCs are not computed in the current model configuration.

 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). 

A.2 Model dynamical parameterizations

 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 giving colder or warmer than average conditions during the winter and spring in each 722 hemisphere. In section 3.6.2, we examine the sensitivity of the HT  $H_2O$  anomaly on the background stratospheric conditions using this modified wave forcing.

 Momentum deposition and vertical eddy diffusion from breaking gravity waves in the stratosphere and mesosphere are computed following the parameterization originally developed 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 ( $\sim$ 325 hPa) as a function of latitude and season, with the slower phase speed waves having larger momentum flux (Holton and Zhu, 1984). The parameterization also solves separately for a single stationary gravity wave generated by flow over orography. Here, vertical profiles of momentum deposition 733 are computed on a longitude-latitude grid ( $10^{\circ}$  x  $4^{\circ}$  grid spacing), with the model zonal mean zonal wind used for each longitude. The orographic surface forcing is based on a multi-year average of monthly zonal gravity wave surface stress from the earlier version 3 of the Whole Atmosphere Community Climate Model (Garcia et al., 2007; see also McFarlane, 1987). 

 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).

A.3 Model QBO simulation

 Simulation of the QBO in equatorial zonal wind is based on previous 2D model studies which parameterize 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).

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
- of the semiannual oscillation in the upper stratosphere and lower mesosphere.
- 

 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 757 range of  $\pm 40$  m/sec at intervals of 2 m/sec (41 waves).

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

 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).

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A.4 Comparison of model tracers with observations

 In this section, we provide an evaluation of the model transport fields by comparing the age of air and H2O simulations with observations. We also show the model ozone response to the HT H2O anomaly compared with MLS observations at SH low-middle latitudes during 2022-2023. 

A.4.1 Age of Air

 Stratospheric mean age of air is a widely used diagnostic that tests the overall fidelity of model 791 transport. Figure A2 shows the age of air derived from measurements of  $SF_6$  and  $CO_2$  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 794 latitudes that may reflect photochemical influences on  $SF_6$  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).

 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 803 important to provide a reasonable simulation of the  $H_2O$  anomaly decay rate (Figure 7).

A.4.2 Water vapor

 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 H2O anomaly (section 2.2), but

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

 The model simulates the full water vapor field generally well in the latitude-height domain compared with MLS version 5 (v5, Figure A3), as well as the time evolution through November 2023 in the mid-stratosphere (Figure A4). The model reproduces the observed transport of large 815  $\text{H}_2\text{O}$  concentrations ( $> 7$  ppm) associated with the HT anomaly from the initial injection to SH 816 mid-high latitudes through November 2023. The H<sub>2</sub>O anomaly appears in the Arctic as a small 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  $\sim 0.5$  ppm throughout much of 819 the stratosphere, likely reflecting the amount of H<sub>2</sub>O entering through the tropical tropopause in the model which is specified from the Upper Atmosphere Research Satellite monthly reference atmosphere (Randel et al., 2001). Some of the model differences with MLS are also due to atmospheric variability not resolved in the simulation. Isolation of the tropical stratosphere 823 below  $\sim$  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). 

826 The model simulates the Antarctic  $H_2O$  distribution generally well compared with MLS (Figure 827 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 September 2023, as well as possible underestimation of ice sedimentation.

832 Both the MLS data and model show a return to more typical low  $H_2O$  values (< 4-5 ppm) in the equatorial mid stratosphere by early 2023 (Figure A4). This reflects the isolation of the tropics from mid-latitudes and the upward transport of drier air from the tropopause, as seen in the equatorial time-height sections (Figure A6a-b). As discussed in section 3.2, the model zonal wind QBO in Figure A6b is in roughly the same phase as the Singapore radiosonde data, with easterlies at 30 hPa at the time of the eruption through mid-2022 ("E" in Figure A6a-b). The corresponding QBO-induced circulation, with relative descending motion over the equator, slows the overall ascent in the tropical mid-stratosphere. As a result, upward transport of the enhanced

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

- 841 in this regard (Figure A6a-b). By late 2022 early 2023, the OBO 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  $H_2O$  plume with large water vapor
- concentrations (> 7 ppm) in the equatorial upper stratosphere through November 2023 in both

MLS and the model.

- 
- 847 A model sensitivity test reveals that this upward transport of the  $H_2O$  plume is somewhat
- dependent on the phase of the QBO at the time of the eruption. With the model QBO in a
- 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
- 853 different phases of the QBO at the time of the eruption can cause a H<sub>2</sub>O variation of  $\pm 1$ -3 ppm in
- 854 the equatorial mid-upper stratosphere, at least during the first  $\sim$ 1½ 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.
- 
- A.4.3 Ozone
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 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- 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 negative ozone anomalies seen in the MLS data averaged over 15°S-40°S (Figure A7b). However, the anomaly magnitudes are generally underestimated in the model, and some of this bias may be due to not including the HT sulfur injection in the simulation. Most of the model anomalies are driven by circulation effects associated with the QBO (Figure A7c). However, the 870 impact of the HT H<sub>2</sub>O anomaly in isolation is quite small, with ozone decreases of at most 0.03







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# **Figure captions**

**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

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

- 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*: de- seasonalized temperature normalized by the standard deviation (color shading). See text for details. The letters show the locations of the time series in Figure 3.

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

 **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



**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)

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

 **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%.

 **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.

 **Figure 8**. Model anomalies of (a) H2O, (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.

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

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),

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

 **Figure 10.** Time-altitude cross-sections of daily model (a) water vapor, (b) temperature, and (c) 1294 ozone anomalies averaged over the Antarctic polar cap ( $65^{\circ}$ S-90 $^{\circ}$ 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; 1298 (b): intervals of  $\pm 0.5$ K and includes the  $\pm 0.1$  and  $\pm 0.2$ K contours; (c) and (d): intervals of  $\pm 1$ DU/km and includes the ±0.2 and ±0.5 DU/km contours.

 **Figure 11.** April-December 2023 daily model anomalies averaged over the Antarctic polar cap (65°S-90°S) at 54 hPa (20 km) for (a) water vapor and ice PSCs, (b) nitrogen species (ppb, solid 1303 lines) and the combined rate of the heterogeneous reaction ClONO<sub>2</sub>+HCl→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 right-hand axis indicates the odd oxygen tendencies in panel (d), and the solar ozone heating rate in

- panel (e).
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1310 **Figure 12.** Model anomalous odd oxygen (O+O<sub>3</sub>) tendencies over the Antarctic polar cap (65°S-1311 90°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).

 **Figure 13.** Daily model total ozone anomalies over the 2022-2031 period. Contour intervals are 1317 -5 DU and include the ±1 DU contours to show minor effects.

 **Figure 14.** Daily total ozone seasonal cycle for (a) 2023 Antarctic (63°S-90°S) and (b) July 2023 - 1320 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.

 **Figure A2.** Age of air at: (a) 20 km (~50 hPa) derived from ER-2 aircraft observations of SF<sup>6</sup> (blue 1330 asterisks) and  $CO<sub>2</sub>$  (red triangles), and (b)-(d) vertical profiles of the age of air derived from 1331 balloon measurements of  $SF_6$  (blue asterisks, green plus signs) and  $CO_2$  (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.

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

 **Figure A4.** Time-latitude cross-sections of daily water vapor (ppm) at 28 hPa for January 2022- 1340 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. 

 **Figure A6.** Time-altitude cross-sections of daily water vapor (ppm) averaged over 10°S-10°S for January 2022-November 2023 from: (a) MLS v5, and model simulations with the zonal wind QBO in the same phase (b) and roughly 180° out of phase (c) with the Singapore radiosonde observations. Panel (d) shows the model difference, (c) minus (b). Maximum easterly ("E") and westerly ("W") zonal winds at 30 hPa are from the Singapore observations in panel (a) and the respective model simulations in panels (b) and (c). The model is forced with the MLS-derived H2O anomaly prior to 1 March 2022 (vertical dotted line in panels (b)-(d)). The contour interval is 1 ppm.

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