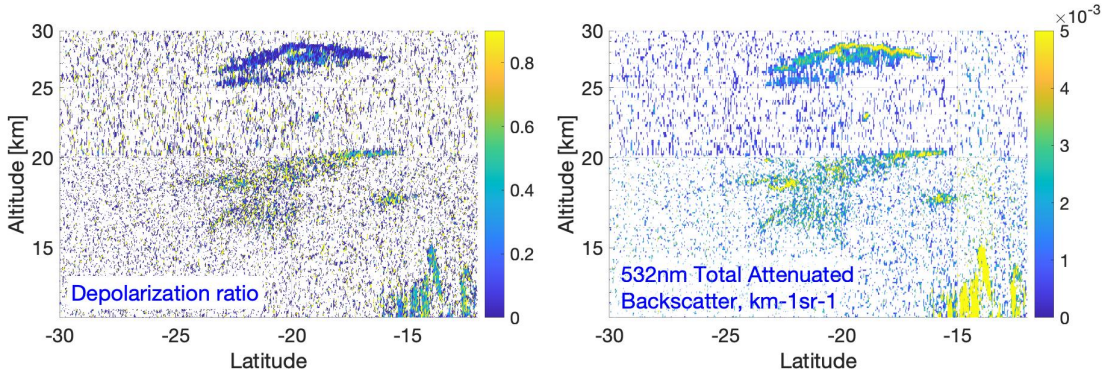
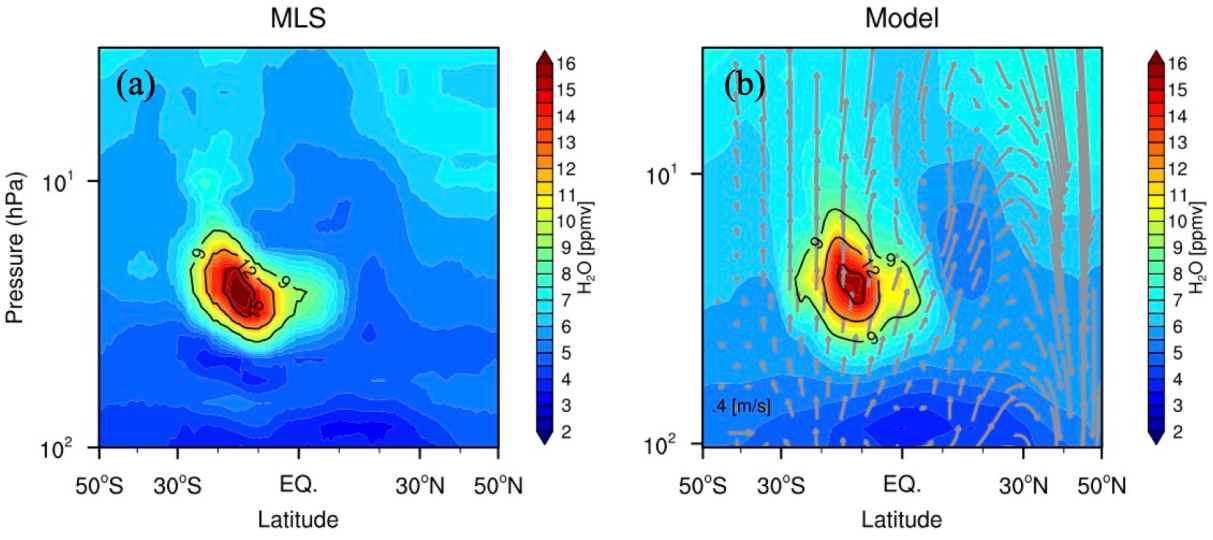
Supplementary Information

Perturbations in stratospheric aerosol evolution due to the water-rich plume of the 2022 Hunga-Tonga eruption

Zhu et al.

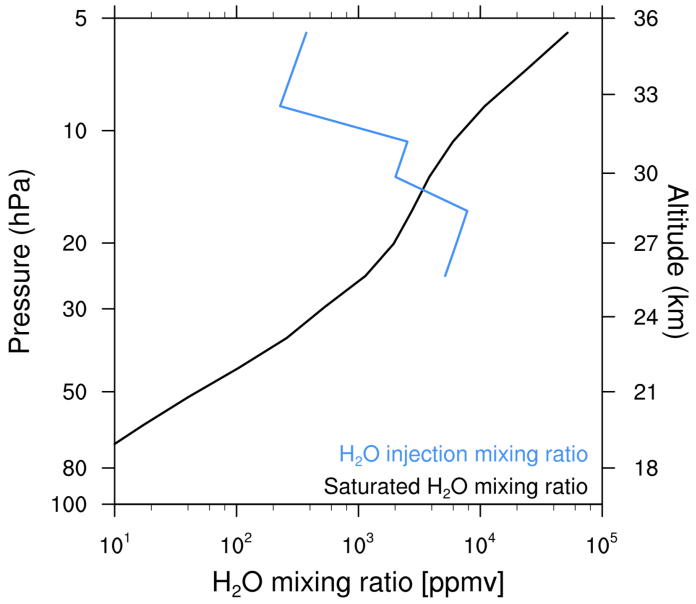


Supplementary Figure 1. The CALIPSO observation on Jan 16. The left panel is the depolarization ratio showing the plume at 30 km with small depolarization and the plume at 20 km with large depolarization. The right panel shows the backscatter coefficient.

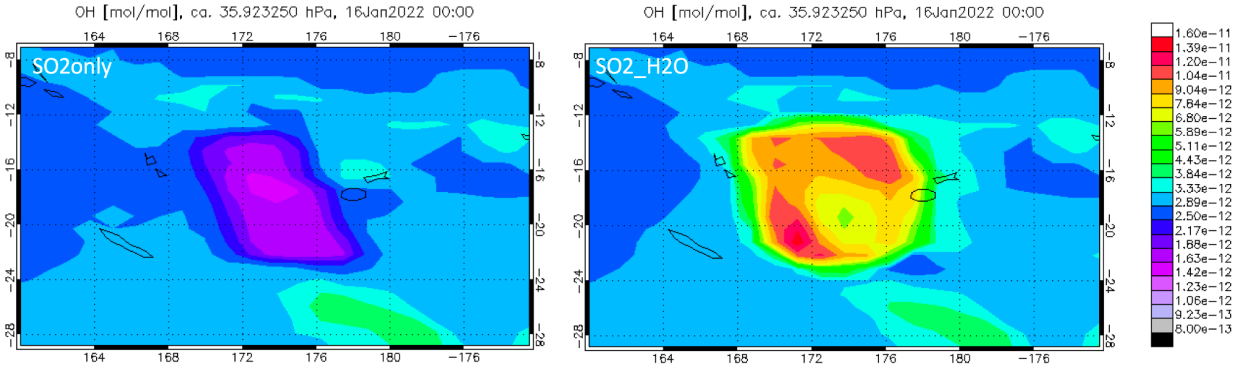


Supplementary Figure 2. Zonal average H2O comparisons between MLS and the model simulation. Zonal average H_2O from MLS v4 data on March 1 (a) and the SO_2_H_2O case with

Brewer-Dobson circulation (vectors) superimposed on March 1 (b). The simulation shows a positive bias in the background because WACCM generally has a wet bias in the stratosphere compared to MLS[1].



Supplementary Figure 3. The H_2O injection scheme relative to the H_2O saturation ratio in the stratosphere. The black line is the saturated H_2O mixing ratio near HTHH on Jan 15 based on the model nudged GEOS5 temperature profile and the temperature profile at the Fiji radiosonde station using the vapor pressure equation of Goff & Gratch[2]. The blue line is the mixing ratio of H_2O injection assuming all H_2O is in the vapor phase in the H2O_SO2 case near HTHH on Jan 15. The figure shows water vapor is saturated at pressures above 15 hPa where ice forms.

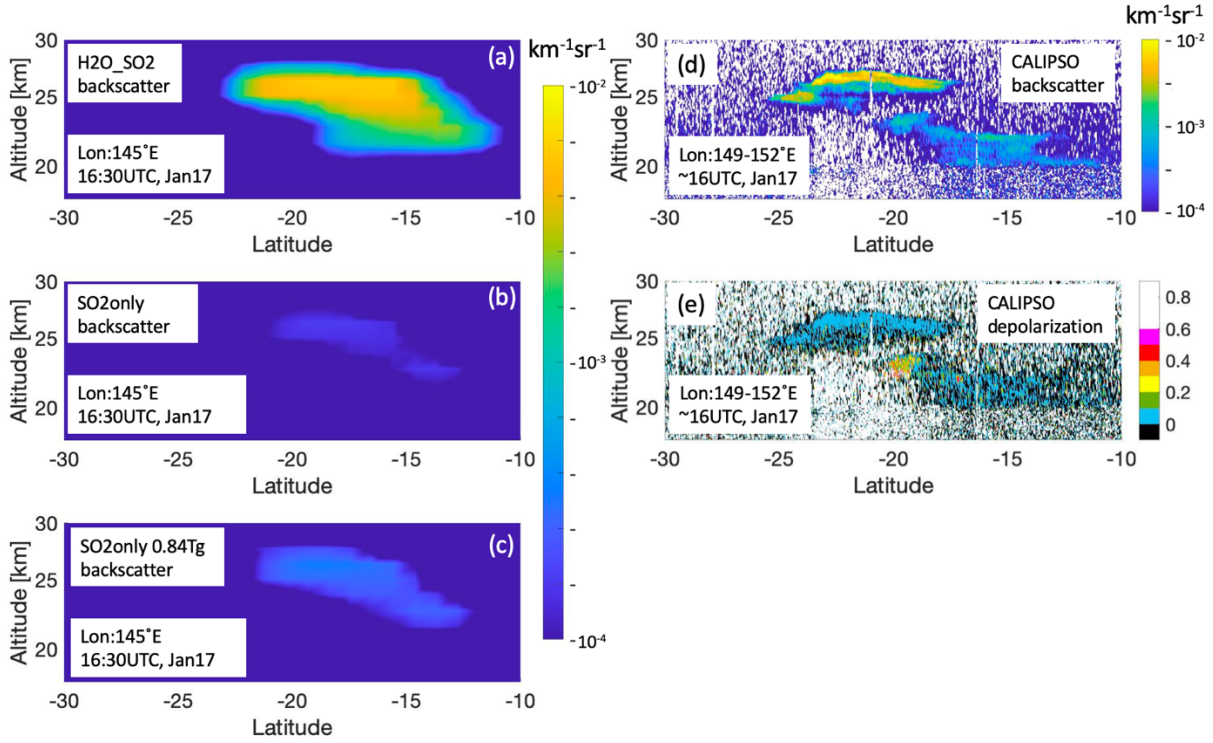


Supplementary Figure 4. The OH concentration at 35 hPa after the eruption. The left panel without water injection shows the OH gets depleted in the SO_2 plume. The right panel with water injection shows H_2O converting to OH rapidly inside the plume.

Supplementary Note 1

In this paragraph, we compare several model simulations with various injection amount and latitude bands to choose the best simulation cases.

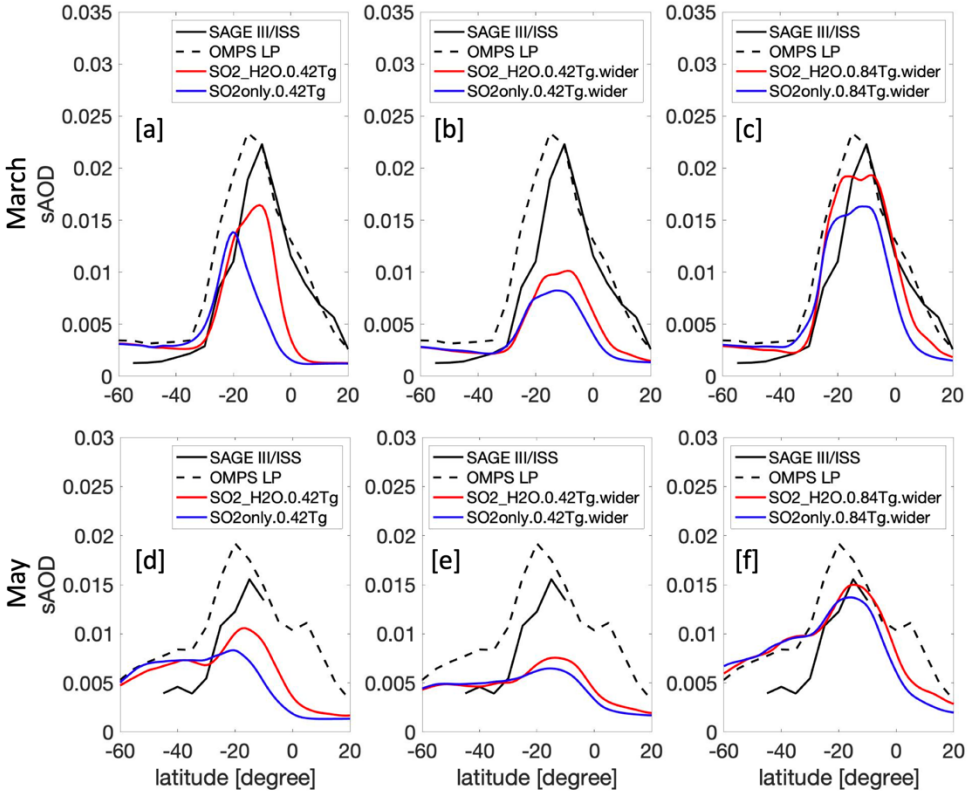
Figure S5b shows that the H2O_SO2 case explains the CALIPSO backscatter. Without water injection, even doubling the injection for the SO2 case cannot create large extinction for the first month as shown in Figure S5c. CALIPSO shows the backscatter peaks about $1e^{-3}$ to $1e^{-2}$ $km^{-1}sr^{-1}$ near 25 km (Figure S5d), which can be reproduced by the SO2_H2O case (Figure S5a). While the SO2only case with 0.42 injection (Figure S5b) only peaks at about $2e^{-4}$ $km^{-1}sr^{-1}$; doubling the SO2only case injection (0.84Tg) peaks at about $4e^{-4}$ $km^{-1}sr^{-1}$ (Figure S5c). Therefore, the SO2_H2O case is better than the SO2only case because the water injection is needed for immediately creating high extinction aerosol layers as observed by both OMPS (Figure 3) and CALIPSO, which is similar to a hypothetical experiment done by LeGrande et al. [3] that simulates the Pinatubo sized eruption with ~150 Tg of water injection.



Supplementary Figure 5. The backscatter coefficient comparisons between several model cases and CALIPSO. CALIOP backscatter coefficient at 532 nm (d) compared with the simulation on Jan 17 ~ 16 UTC along CALIOP track: a) 0.42 Tg SO2_H2O case, b) 0.42 Tg SO2only case and c) 0.84 Tg SO2only case. Simulated aerosol backscatter is calculated using the simulated visible extinction coefficient divided by the extinction-to-backscatter ratio (value = 55 sr) for volcanic sulfate suggested by the Wyoming balloon particle counter data calculation two years after the Mt. Pinatubo eruption in 1993 (value ~ 50 sr)[4,5] and CALIOP lidar ratio retrievals for sulfate-rich aerosol layer (value = 60 sr)[6]. e) CALIOP depolarization ratio shows low

depolarization for both plumes, but the lower plume shows a small segment of high depolarization near 23 km and 20˚S.

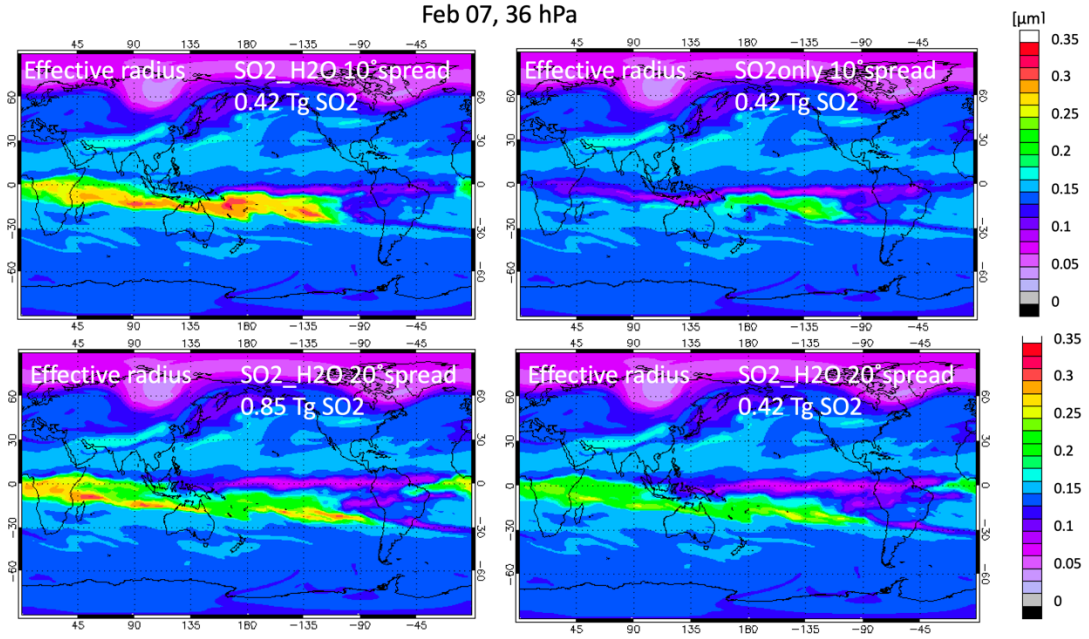
Another issue with the model is that the area covered by the cloud is larger than in the model. Figure S6 shows sAOD averaged over March and May from SAGE III/ISS and OMPS. SO2_H2O cases always produce higher sAOD than the SO2only cases for the same injection scenarios. If we injection in a 10-degree latitude band (S6a and S6d) with 0.42 Tg of SO_2, we get a smaller sAOD near tropics and NH, but SH mid-latitude compares with OMPS well. If we injection in a 20-degree latitude band (S6b and S6e) with 0.42 Tg of SO_2, the sAOD is significantly reduced. The model matches the observation the best when injecting 0.84 Tg of SO_2 into 20-degree latitude bands (S6c and S6f).



Supplementary Figure 6. zonal average sAOD in March and May from OMPS, SAGE III-ISS, and several model cases. The black line is from SAGE III/ISS, the dashed black line is from OMPS LP. The red color is SO2_H2O cases with various injection amounts and injection widths; the blue color is the SO2only case. In panels b, c, e, and f, the modeled injection latitude bands double the original injection latitude bands.

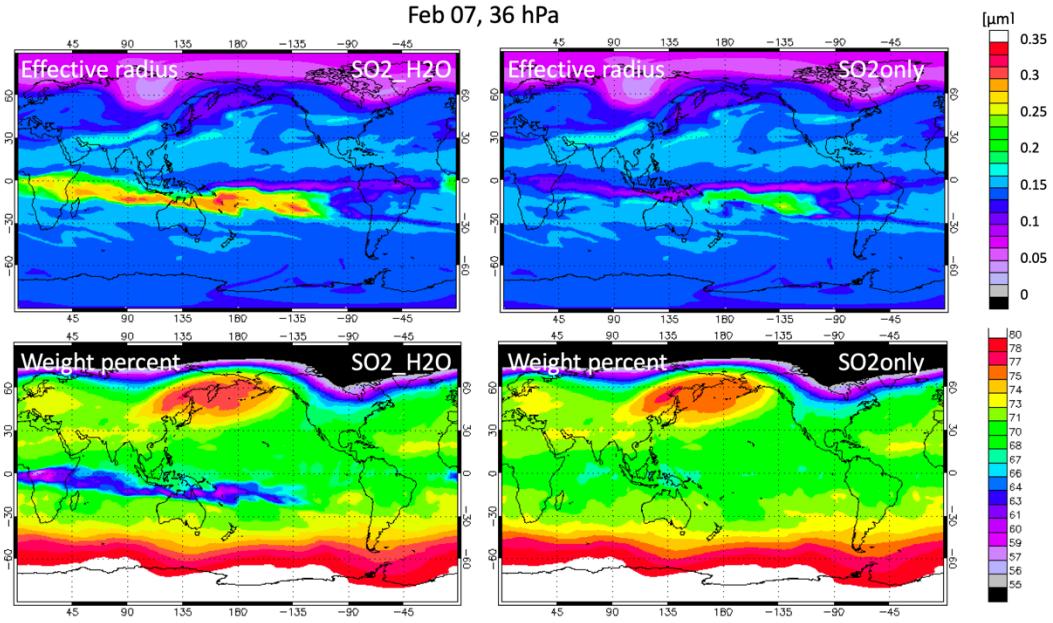
Artificially injecting into a 20-degree band can cause changes in the initial microphysics, resulting in smaller effective radii of particles and smaller optical depths. As stated in Murphy et al.[7], sulfate particles with a radius around 0.3 µm give the largest extinction for a given mass. This can be one of the reasons why we need to double the injection to produce observed sAOD. Figure S7 shows the effective radius from several model runs. We can see the SO2_H2O case has the effective radius of 0.3 µm. If we spread the plume wider, the effective radius is reduced to 0.2 µm.

If we spread the plume wider and increase the injection to 0.84 Tg, the effective radius is closer to 0.3 µm. OMPS clearly shows the plume spreading fast from 10 degrees wide to 30 degrees wide during the first couple of weeks, which could be caused by localized sub-grid winds due to the volcanic debris that the model does not have the spatial resolution to simulate.

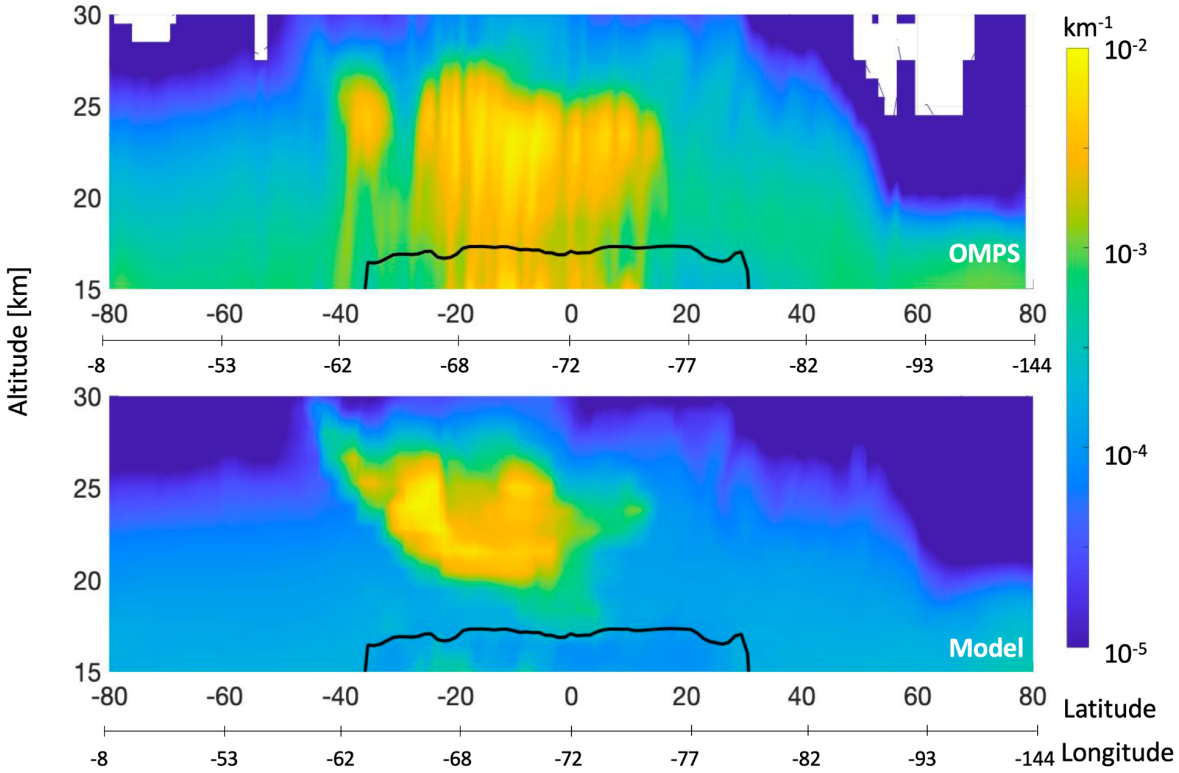


Supplementary Figure 7. The effective radius of the particles from several model runs.

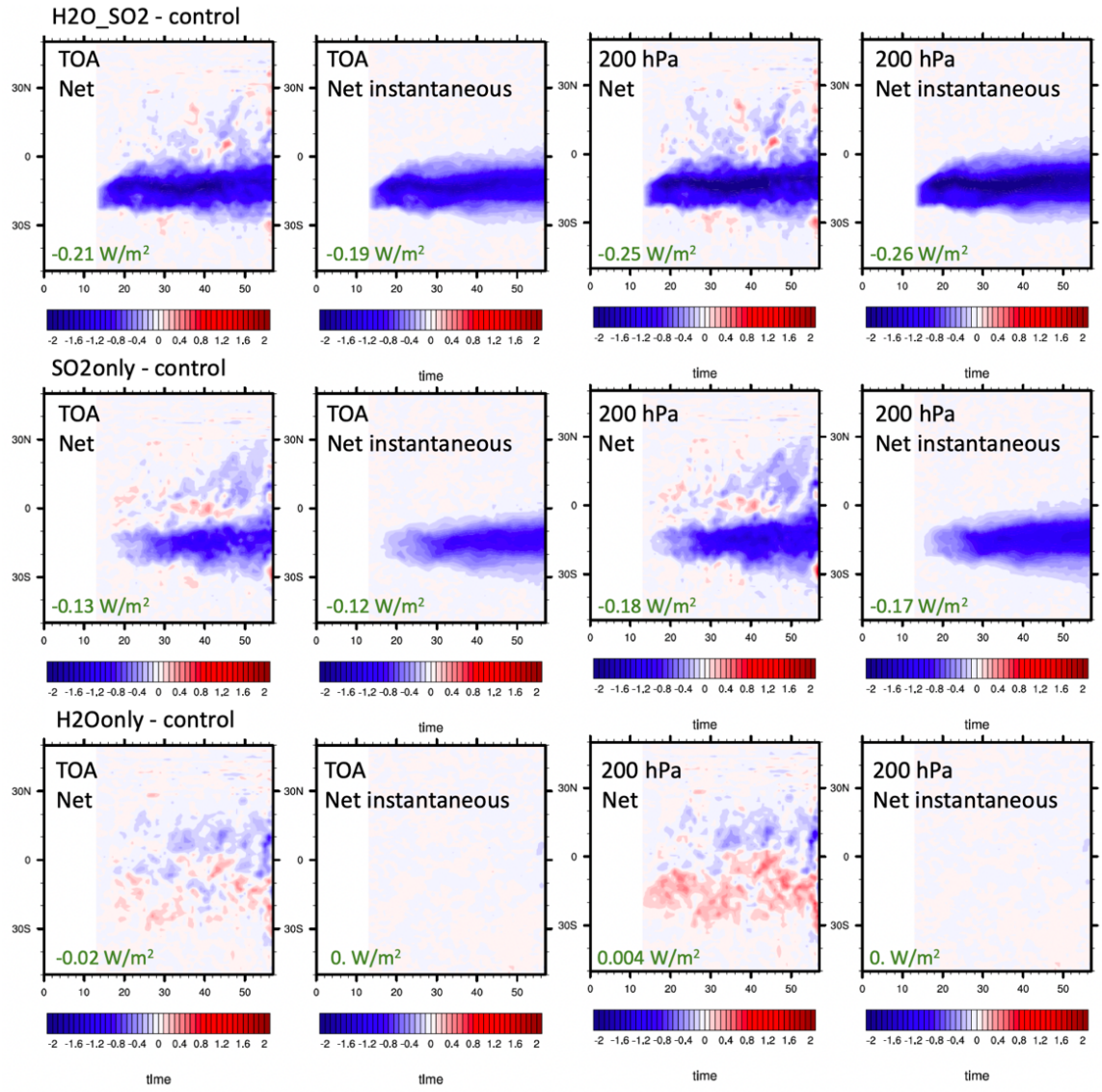
Based on our current best knowledge of SO_2 injection: Carn et al.[8] summarized the estimation as about 0.4-0.5 Tg. However, our model produces the best match to optical data with the 0.84 Tg injection with 20-degree latitude band injection. Our current model setting is the best estimation of injection area and injection amount of SO_2 and H_2O based on observations. However, there could be ways to remove this conflict in the future. For example, small ash particles coated with sulfate could increase the extinction for a smaller SO_2 injection and still be round allowing them to not depolarize and conflict with CALIPSO data. Ash particles can have a small density which allows particles to be moderately large and still persist without falling[9].



Supplementary Figure 8. The effective radius and weight percentage of sulfate aerosol ($SO4^{2-}$: ($SO4^{2-}$+H_2O) inside the particle) comparisons between the SO2_H2O case and the SO2only case on February 7 at 36 hPa.

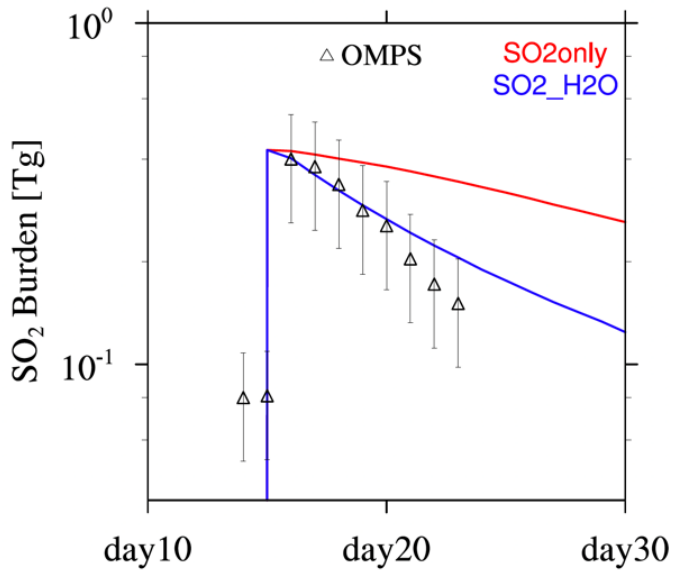


Supplementary Figure 9. One of the OMPS-LP extinction profiles on March 2 compared with the SO2_H2O simulation for the same sampling. They show a consistent vertical extension above 20 km. OMPS data near the tropical tropopause (black line) may be affected by tropical cirrus clouds because this orbit data hasn't had cloud screening applied. They both show that volcanic sulfate extends from 40˚S to 10˚N, but the simulation has smaller extinction values in the northern hemisphere.



Supplementary Figure 10. The net radiative effect anomaly from three model cases: H2O_SO2, SO2only, and H2Oonly minus control. The first column shows the TOA adjusted radiative effect. The second column shows the TOA instantaneous aerosol effect. The third column shows the adjusted radiative effect at 200 hPa. The fourth column shows the instantaneous aerosol effect at 200 hPa. The adjusted radiative effect includes flux changes directly caused by a change in atmospheric composition and resulting from changing atmospheric or surface state. The adjusted radiative effect is calculated using the clear sky (no cloud) net flux. The instantaneous aerosol effect is calculated using the clear sky (no cloud) net flux minus the clean sky (not cloud and no aerosol) net flux. The global mean values for each panel are marked in the bottom left corner. The instantaneous aerosol effect shows aerosol gives negative radiative effects comparable to the adjusted radiative effect. The radiative effect from water is positive in the Southern hemisphere but is much smaller than that of the aerosol in both TOA and 200 hPa. Also, the global averaged radiative effect in the H2Oonly case at TOA in the Southern hemisphere is cancelled out by the negative values in the Northern hemisphere,

resulting in a slightly negative global average value. The instantaneous effect is small in the H2Oonly case because no volcanic aerosol is injected.



Supplementary Figure 11. The time series of the stratospheric SO_2 burden similar to Figure 2. The triangles are SO_2 measurements by OMPS-NM without applying the detection limit. The solid lines are the model simulations without applying the detection limit.

Data Availability

The supplementary data are available at (https://osf.io/6ZXFV/) with a permanent doi 10.17605/OSF.IO/6ZXFV.

Supplementary References

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