

Overview

Simulating a volcanic eruption's impact on stratospheric aerosol presumes knowledge of its injection height and mass, plume composition, location, and atmospheric state variables. In situ measurements are key to validating these assumptions. Using measurements from the Tonga Rapid Response Experiment (TR²Ex) and subsequent Balloon Baseline Stratospheric Aerosol Profiles (B²SAP) soundings, we track the evolution of the aerosol and water vapor plumes following the Hunga Tonga-Hunga Ha'apai (HT-HH) eruptions on January 13 and 15, 2022.

Together, the HT-HH eruptions (21 °S, 175 °W) are estimated to have injected very large quantities of water vapor (>100 Tg H₂O) paired with a modest amount of sulfur dioxide (~ 0.49 - 0.56 Tg SO₂; 0.25 - 0.28 Tg S) into the stratosphere. Whereas the typical e-folding lifetime of SO₂ in the stratosphere is approximately one month, with its conversion gradually increasing the amount of sulfuric acid (H₂SO₄) aerosol, we observed large perturbations to stratospheric aerosol within a few days of the HT-HH eruption. Aerosol layers with the highest mass corresponded to regions of the plume with high water vapor mixing ratios. Assuming that particles were composed of H₂SO₄, in situ measurements of aerosol number and size can be used to determine vertically integrated aerosol mass and ambient stratospheric aerosol optical depth (sAOD). Together with OMPS sAOD (λ = 997 nm) and MLS H₂O data, these in situ observations from La Réunion (21 °S, 55 °E) were used to calculate the total aerosol sulfur (S) mass (0.22 Tg S after 11 days) of the young plume and provide an estimate of the stratospheric SO₂ e-folding lifetime in these wet conditions (~ 7.3 days), consistent with the initial estimate of SO₂, the observed partitioning of S between TR²Ex SO₂ (~20 %) and H₂SO₄ aerosol (~80 %), and recent modeling work. B²SAP water vapor and particle size distribution measurements from regular soundings from Lauder, NZ, Hilo, Hawaii, and Boulder, CO, as well as La Réunion in the subsequent months provide compelling evidence of gravitational sorting and settling of aerosol, as well as dilution and transport to higher latitudes in both hemispheres of the water vapor and aerosol plumes.

The Payloads

Instruments:
 The Portable Optical Particle Spectrometer (POPS) number and size (D_p >140 nm)

Cryogenic frost point hygrometer instruments (CFH) on TR²Ex and NOAA frost point hygrometer (FPH) on B²SAP flights

SO₂ sonde measurements (TR²Ex)

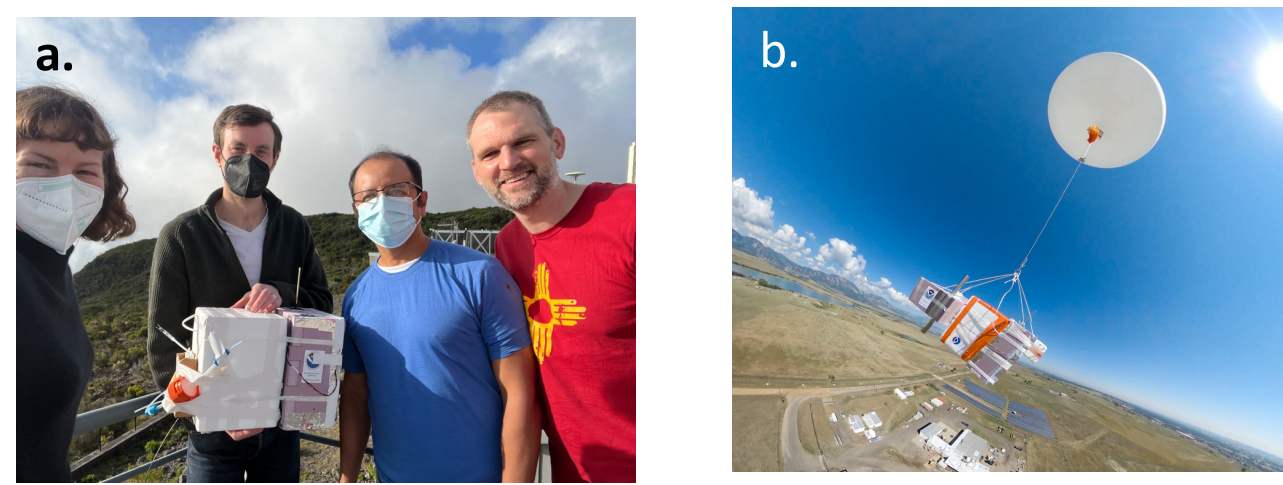


Figure 1. Photo of a TR²Ex payload with a Portable Optical Particle Spectrometer (POPS; Gao et al. 2016) & SO₂ sonde (Yoon et al., 2021), (ozonesonde & CFH TR²Ex payload not shown (a) and a B²SAP payload with a NOAA FPH (Hall et al., 2016), POPS, and ozonesonde.

Site Descriptions

Sites with regular B²SAP (4+ yearly) launches are represented with stars, whereas sites of B²SAP Intensive Operations Period (IOP) measurements are shown as points.

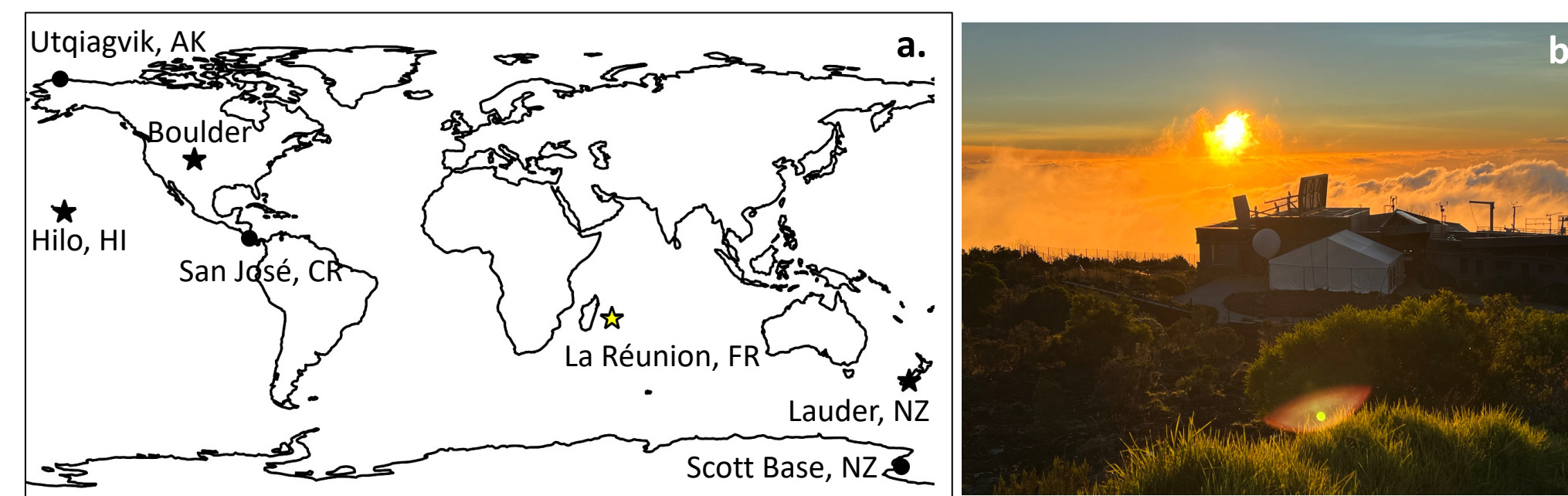


Figure 2. Map of B²SAP sites (a) and photo of the Maïdo observatory on La Réunion (b).

Immediate impact on stratospheric aerosol and water vapor

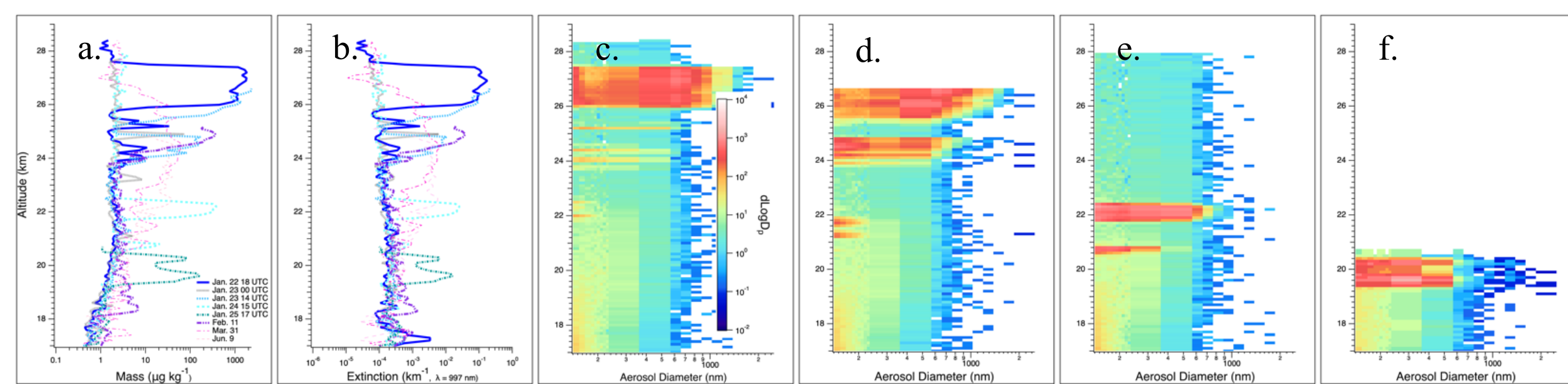


Figure 3. Vertical profiles of aerosol dry mass and extinction between Jan. and Jun. 2022 (a-b); calculated from POPS size distribution data and vertical profiles of measured aerosol size distributions during TR²Ex (c-f).

Due to wind shear, the plume was stretched into a thin, slanted aerosol layer. This layer was sampled multiple times within 6-10 days of the eruption on Jan. 15.

Size distributions reveal an abundance of larger particles (400 nm - > 1 μm) in parts of the plume above 25 km, corresponding to a region of the plume with substantial water vapor enhancements.

The aerosol composition is unknown - assumed to be sulfate ()

Balloon-borne water vapor sondes recorded > 300 ppmv H₂O in the upper altitude part of the plume above 25 km or Jan. 22, 2022 at 20 S. Mixing ratios in the lower altitude of the plume were 10x smaller.

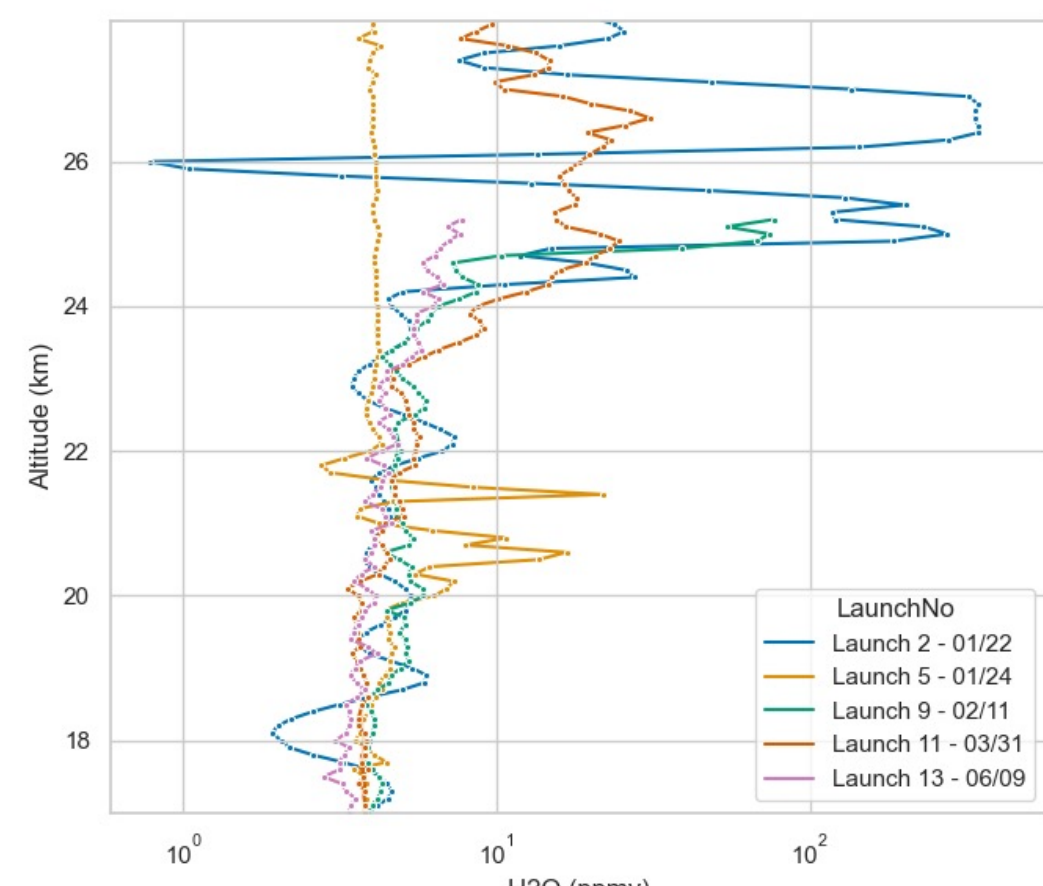


Figure 4. Vertical profiles of stratospheric water vapor measurements from cryogenic frost point hygrometer (CFH) and NOAA frost point hygrometer (FPH) instruments.

Aerosol extinction and mass, as well as water vapor mixing ratios remained elevated in the higher altitude part of the plume through Jun. 2022.

Less SO₂ than aerosol in the fresh HT-HH plume

Details:

Aerosol presumed to be composed of sulfate or H₂SO₄. Estimated H₂SO₄ in the plume is abbreviated as eH₂SO₄.

$$eH_2SO_4 = \sum n \cdot \frac{4}{3} \pi (D_p/2)^3 \cdot \rho_{(H_2SO_4)} / MW_{(S \text{ in } H_2SO_4)}$$

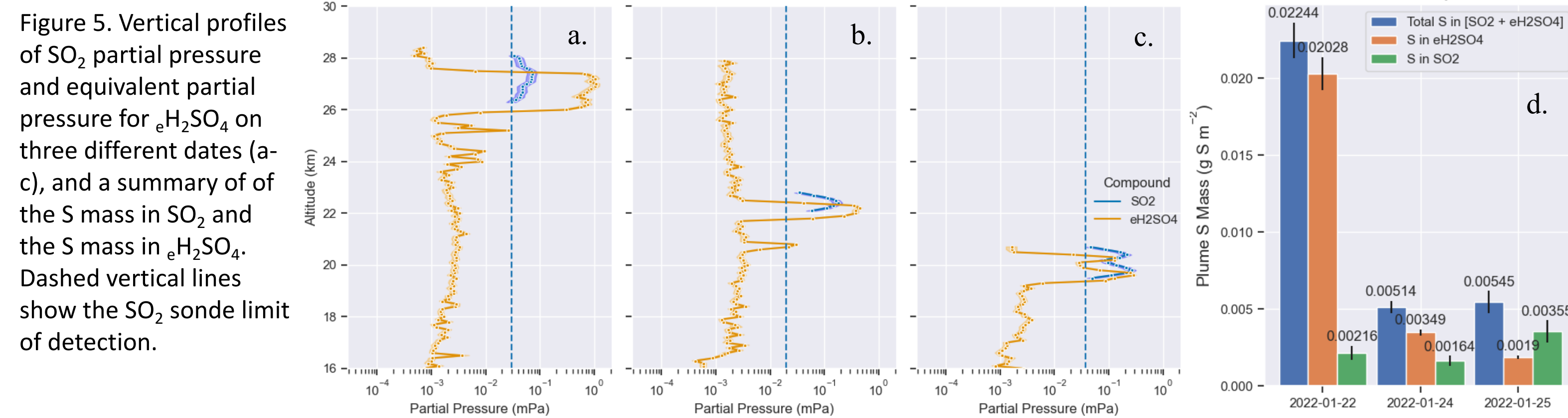


Figure 5. Vertical profiles of SO₂ partial pressure and equivalent partial pressure for eH₂SO₄ on three different dates (a-c), and a summary of the S mass in SO₂ and the S mass in eH₂SO₄. Dashed vertical lines show the SO₂ sonde limit of detection.

Large H₂O enhancements can speed up SO₂ oxidation by increasing OH availability (e.g., in the HT-HH plume; Zhu et al., 2020, 2022)

Leveraging satellite global coverage and in situ measurements to calculate the aerosol layer mass

Suomi National Polar-Orbiting Partnership (SNPP)
 Ozone Mapping and Profile Suite Limb Profiler (OMPS-LP; Taha et al., 2021)



$$\text{Scale: Aerosol column mass (g m}^{-2}\text{)} = 0.139 \text{ sAOD} + 0.001$$

POPS calculated sAOD vs. aerosol column mass Jan. 22 - June. 9 (r² = 0.98, p < 0.001) applied to OMPS-LP sAOD to calculate total aerosol mass.

Figure 6. Histogram of (log₁₀) daily OMPS-LP sAOD data (λ = 997 nm) for Jan. 10 between 50 N and 50 S, prior to the HT-HH eruption and daily OMPS-LP data for Jan. 22 (a). The maximum sAOD value from Jan. 10 data (black line) was used to determine measurements made within and outside of the plume thereafter. The median of data on Jan. 22 (red line), used as the background sAOD, and three mean absolute deviations above the median (blue line), used to identify outliers, are also shown. Contour plots of eH₂SO₄ aerosol column (g m⁻²), calculated using the equation above and OMPS-LP retrievals of sAOD, for five days from Jan. 18 to Jan. 26 (b-f). On Jan. 22, the gray shaded area shows the maximum plume extent between Jan. 21 - Jan. 23 as determined from MLS H₂O anomalies at 21 hPa is also shown. Locations of HT-HH eruption and La Réunion are marked with a cross and a star, respectively.

Stratospheric lifetime (τ) of SO₂ can be calculated using a linear regression against time

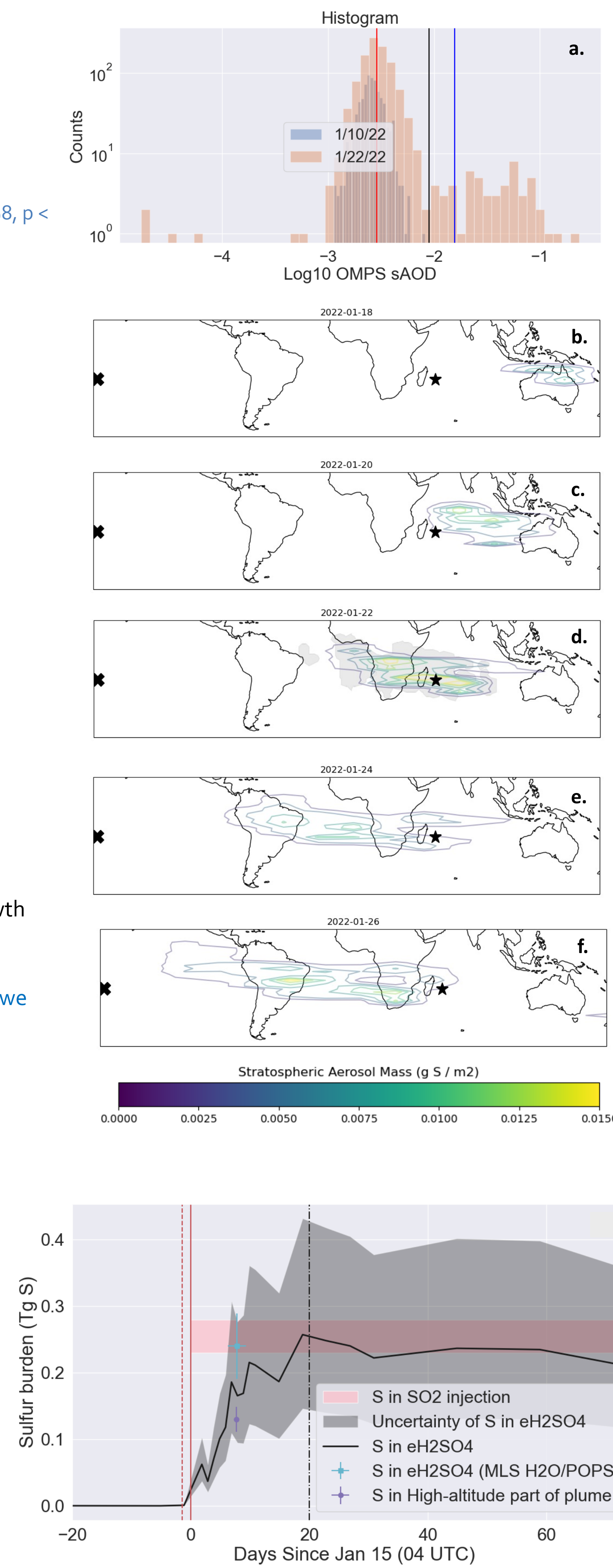
Assumptions:

- SO₂ injection of 0.26 - 0.28 Tg S
- Initial burden of H₂SO₄ ~ 0.005 Tg
- All the SO₂ is converted to H₂SO₄ (not other compounds)
- Ignores potential time lag for particle formation or aerosol growth to diameter ≥ 0.14 μm

• If some of the aerosol was actually ash (e.g., <= 0.012 Tg) ...or if we adopted a more conservative estimate of plume area, the calculated τ_{SO2} would be longer (9-13 days)

Aerosol formation appeared complete within 20 days of Jan. 15 (22 days of Jan. 13)
 The total aerosol layer burden reached ~ 0.26 Tg S (0.52 Tg SO₂ equivalent)
 Second estimate (MLS H₂O) consistent with a modest S injection
 High altitude part of the plume contained the 3/4 of the total S mass in the aerosol layer

Figure 7. A time series of the S burden in eH₂SO₄ aerosol, calculated using a combination of measurements by POPS, OMPS-LP and MLS. The black shaded region is the total calculated uncertainty in the S burden, the pink shaded region is the estimate of the S injected as SO₂ from satellites, the MLS-based element is shown in cyan, and the mass of the plume west of La Réunion on 1/23 0 UTC - vertical profiles indicate that up until this point the plume was observed above 25 km. Dashed vertical lines depict the period of eH₂SO₄ accumulation.



Hygroscopic growth due to H₂O enhancements

Large H₂O enhancements were observed in high-altitude parts of the plume (above 25 km; Fig. 4)

H₂SO₄ aerosol is hygroscopic, which could result in an increase in aerosol diameter of ~ 10% at 500 nm, according to kappa-Köhler theory

Water vapor partial pressure with respect to ice was calculated using the Goff-Gratch equation (1984) from NOAA FPH or CFH frost point temperature.

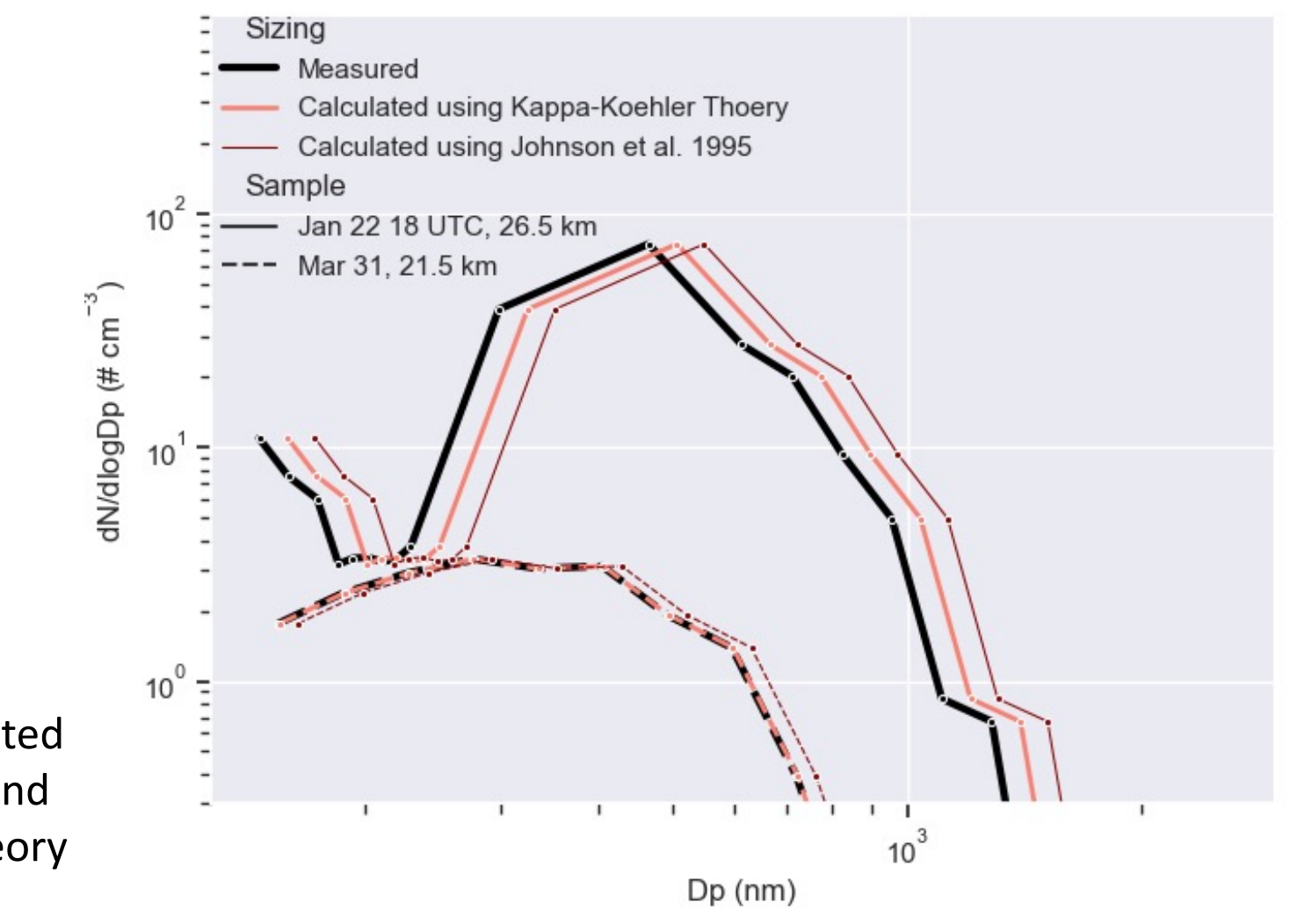


Figure 8. Measured POPS size distributions and calculated ambient size distributions on Jan. 22, 2022 (26.5 km) and March 31, 2022 (21.5 km) using both kappa-Köhler theory and the Jonsson et al. formulation.

Transport and diffusion of the H₂O plume

Good agreement exists between NOAA FPH measurements and MLS retrievals of stratospheric H₂O at sites in both the NH and SH tropics and midlatitudes.

After La Réunion, enhancements in H₂O mixing ratios were first observed in the NH tropics in Hilo, HI.

By May (4 months after the eruption), enhancements in H₂O mixing ratios were observed in Lauder, NZ, and by Jan. 2023 in Boulder, CO (1 year after the eruption)

In the SH and NH midlatitudes, enhancements in H₂O mixing ratios were observed at lower altitudes (and correspondingly lower potential temperatures) than in the tropics, consistent with Brewer Dobson circulation patterns.

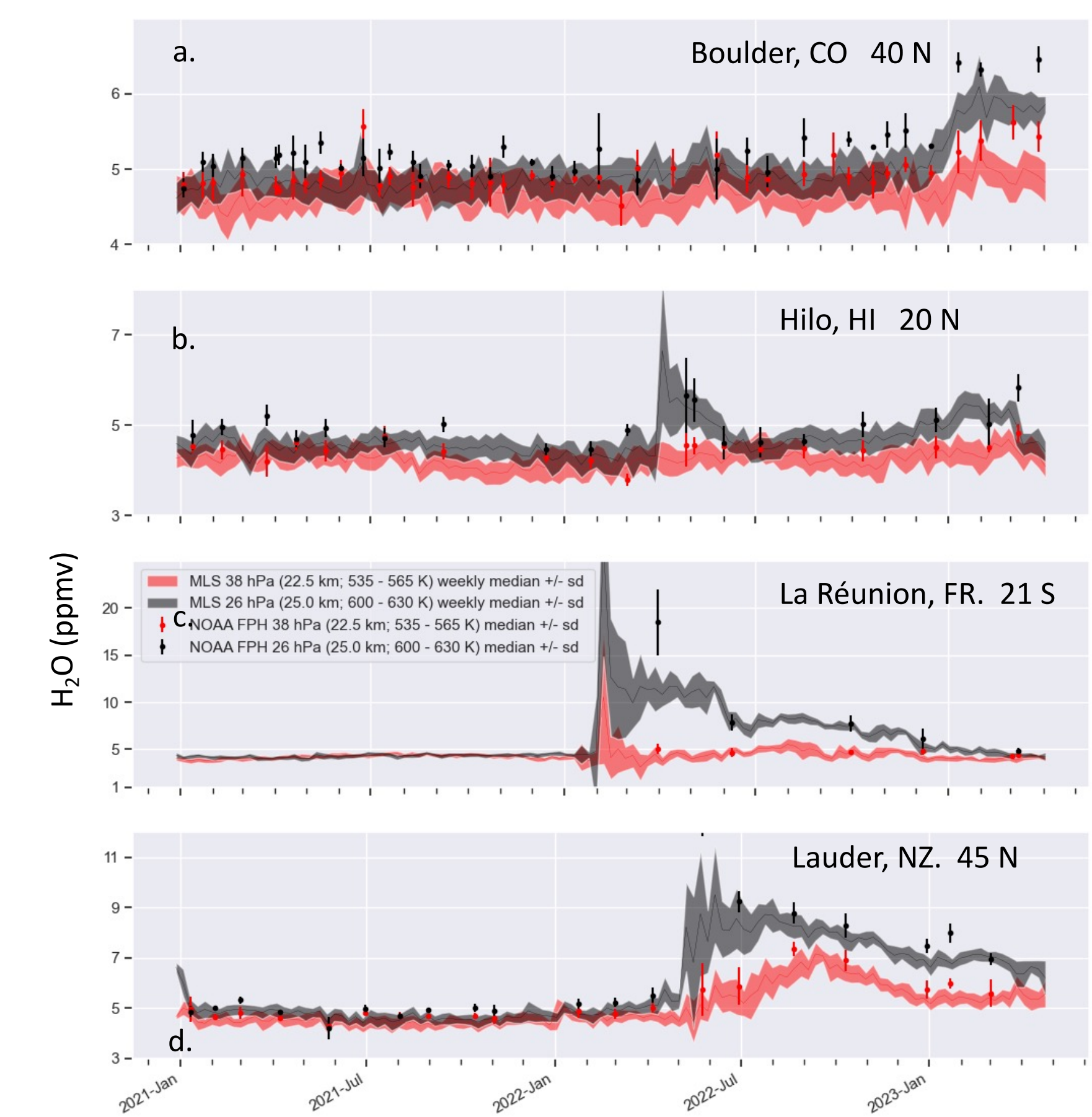


Figure 9. Timeseries of MLS weekly median and NOAA FPH daily median H₂O mixing ratios at two different pressure levels (with their equivalent altitudes and potential temperature ranges) in the upper stratosphere.

Summary

- Observed unexpected abundance of eH₂SO₄ relative to SO₂ in the fresh HT-HH plume.
- With OMPS-LP aerosol retrievals of sAOD, in situ measurements may be used to calculate the total mass of the aerosol plume and constrain the stratospheric lifetime of SO₂
- The calculated mass for HT-HH was ~ 0.25 Tg S and lifetime of SO₂ was ~ 6-9 days (slightly longer if assumptions relaxed)
- Water vapor sondes recorded H₂O mixing ratios of >300 ppmv in the fresh plume in the higher altitude part of the plume (above 25 km)
- Suspect that H₂O enhancements resulted in the hygroscopic swelling of aerosol in the higher altitude part of the aerosol layer
- Following dilution and transport of the H₂O plume to the NH and higher latitudes in the SH, peak H₂O mixing ratios of > 6 ppmv at 20 N (Hilo, HI) in March, 2022, > 9 ppmv at 45S (Lauder, NZ) in May, 2022, > 6 ppmv at 40N (Boulder, CO) in January, 2023.

References/ Acknowledgements

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