Supporting Information for "Strong Evidence of Heterogeneous Processing on Stratospheric Sulfate Aerosol in the Extrapolar Southern Hemisphere Following the 2022 Hunga Tonga-Hunga Ha'apai Eruption"

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1. Figures S1 to S5

Introduction

Supporting Information contains additional figures supplementing the discussion in the main text. Figure S1 shows the reactive uptake coefficients as a function of temperature for the heterogeneous reactions R1–R5 discussed in the main text. The reactive uptake coefficient (or surface reaction probability, γ) is the fraction of collisions between a gas-

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phase reactant and a particle surface that lead to reactive uptake of the trace gas species on/in the particle. Figure S2 shows anomalies of aerosol cross section from the Suomi-NPP Ozone Mapping and Profiler Suite Limb Profiler (OMPS-LP) and of water vapor, HNO_3, ClO, HCl, N_2O and ozone from the Aura Microwave Limb Sounder (MLS) similar to those shown in the main text. Here, however, to facilitate comparison of changes across different quantities and equivalent latitude bands, all anomalies have been normalized by dividing the raw magnitudes by their respective maximum absolute values. Results are shown for the Southern Hemisphere (SH) midlatitudes, subtropics, and tropics. Figure S3, similar to Figures 3 and 8 in the main text, gives the vertical view of the anomalies in various quantities for the SH subtropics. Figure S4 shows MLS HNO_3/N_2O, HCl/N_2O, and O_3/N_2O correlations as in Figures 4 and 9 in the main text, but for the SH subtropics. Figure S5 shows a potential temperature / time section similar to those in Figure S3, but for the anomaly in MLS monthly HO_2 in the SH tropics; in this case, the lowest potential temperature shown is 670 K, as the MLS HO_2 data are not recommended for scientific study at retrieval levels below (i.e., pressures greater than) 10 hPa (Livesey et al., 2020).

References

Burkholder, J. B., et al. (2019). *Chemical kinetics and photochemical data for use in atmospheric studies, evaluation number 19* (Tech. Rep. No. JPL Publication 19-5). Pasadena: Jet Propulsion Laboratory. (Available at http://jpldataeval.jpl.nasa.gov)

Livesey, N. J., et al. (2020). *Version 5.0x Level 2 and 3 data quality and description document* (Tech. Rep. No. JPL D-105336 Rev. A). Pasadena: Jet Propulsion Laboratory. (Available at http://mls.jpl.nasa.gov.)

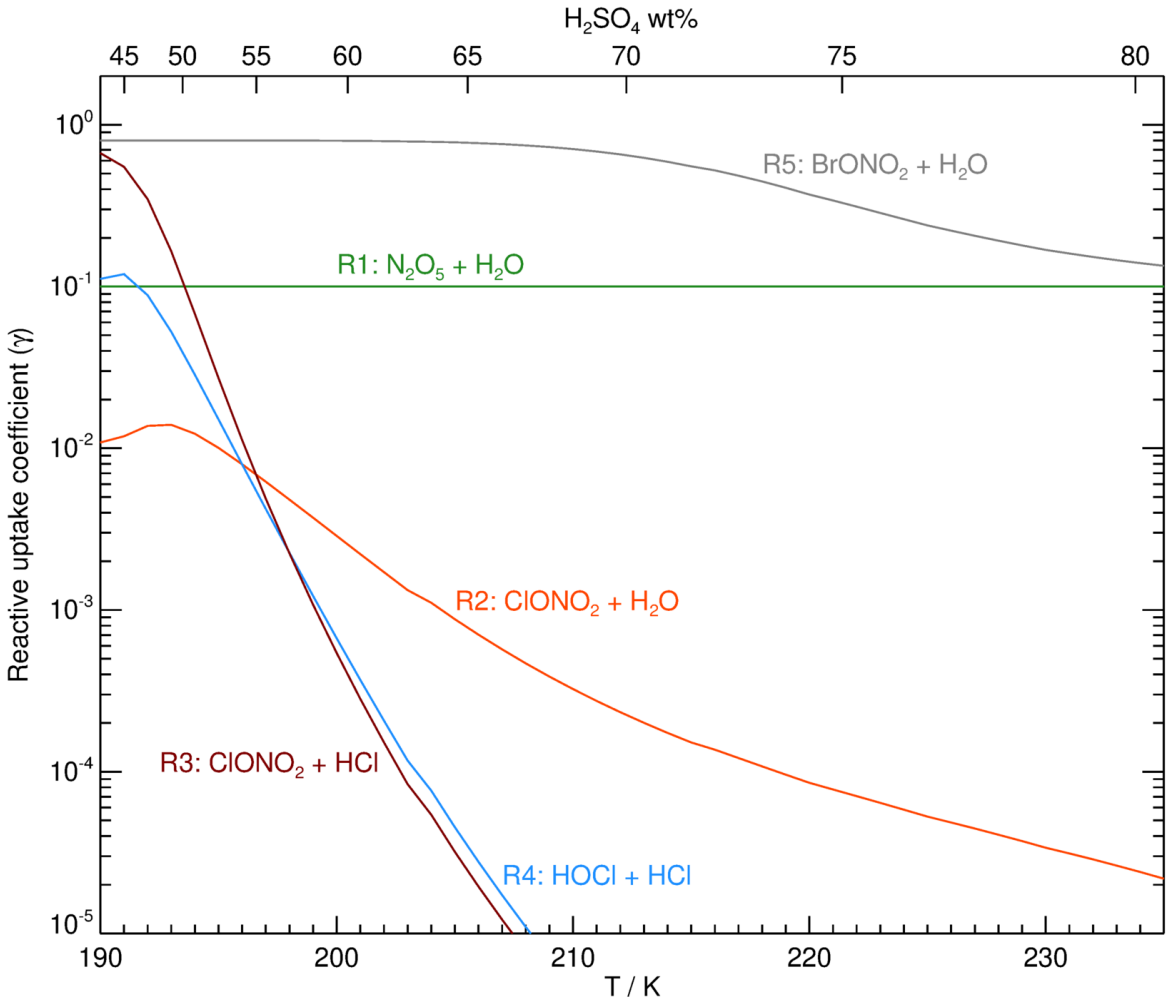

Figure S1. Reactive uptake coefficients (i.e., γ values, a measure of the probability of occurrence of a given reaction) for R1–R5 on stratospheric sulfuric acid aerosols as a function of temperature. Calculations follow the recommendations given by Burkholder et al. (2019) and assume fixed abundances of 5 ppmv H_2O, 1 ppbv HCl, 0.6 ppbv $ClONO_2$, 0.1 ppbv HOCl, and 0.1 ppbv H_2SO_4 for a mean particle radius of 0.1 μm at 50 hPa.

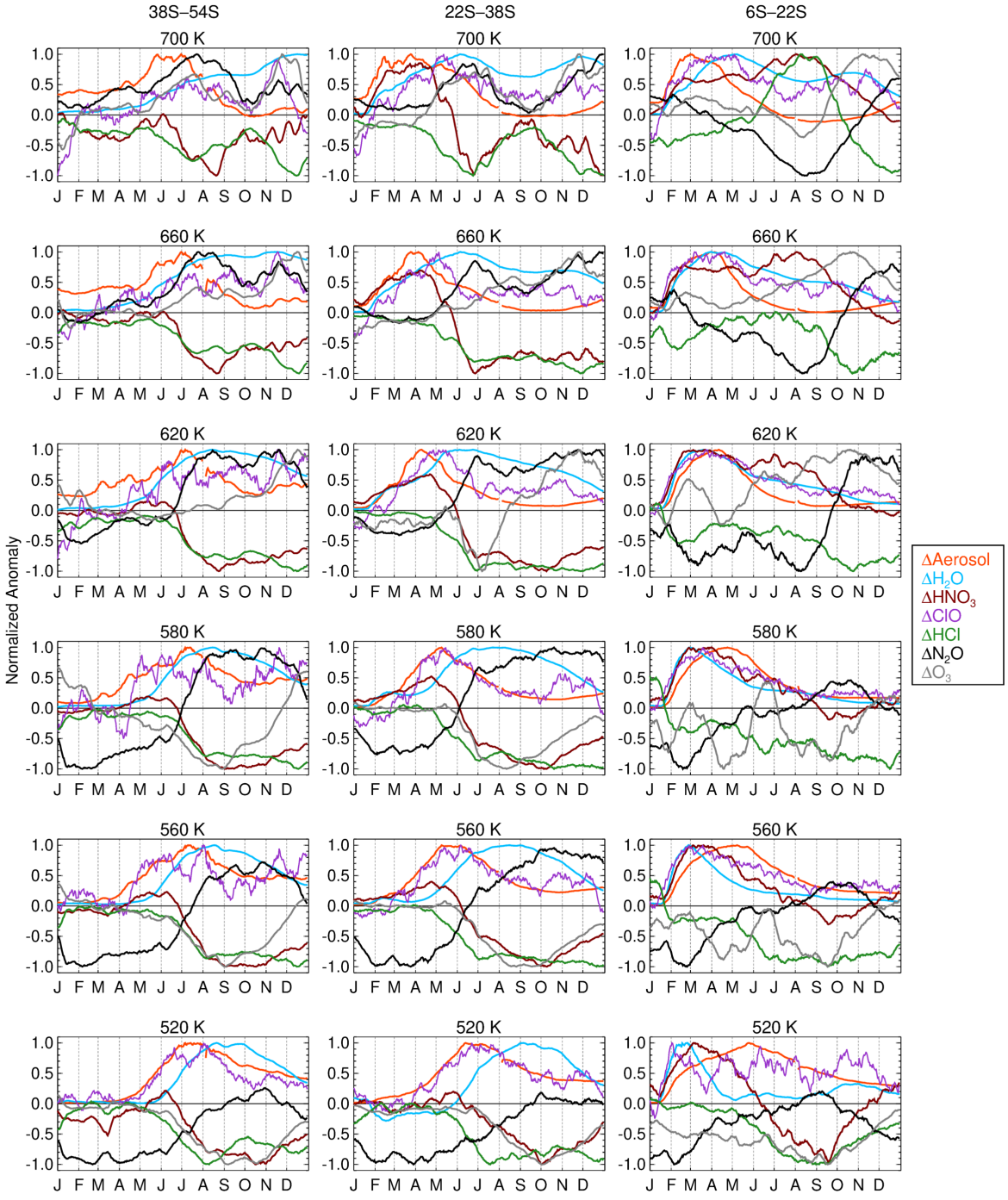

Figure S2. Normalized anomalies of NASA OMPS-LP aerosol cross section at 869 nm and MLS H_2O, HNO_3, ClO (day minus night), HCl, N_2O, and O_3 (see legend) at six selected potential temperatures for the (left) 38°S–54°S, (middle) 22°S–38°S, and (right) 6°S–22°S EqL bands. Anomalies are normalized by dividing the raw magnitudes by their respective maximum absolute values. Solid black horizontal lines mark zero.

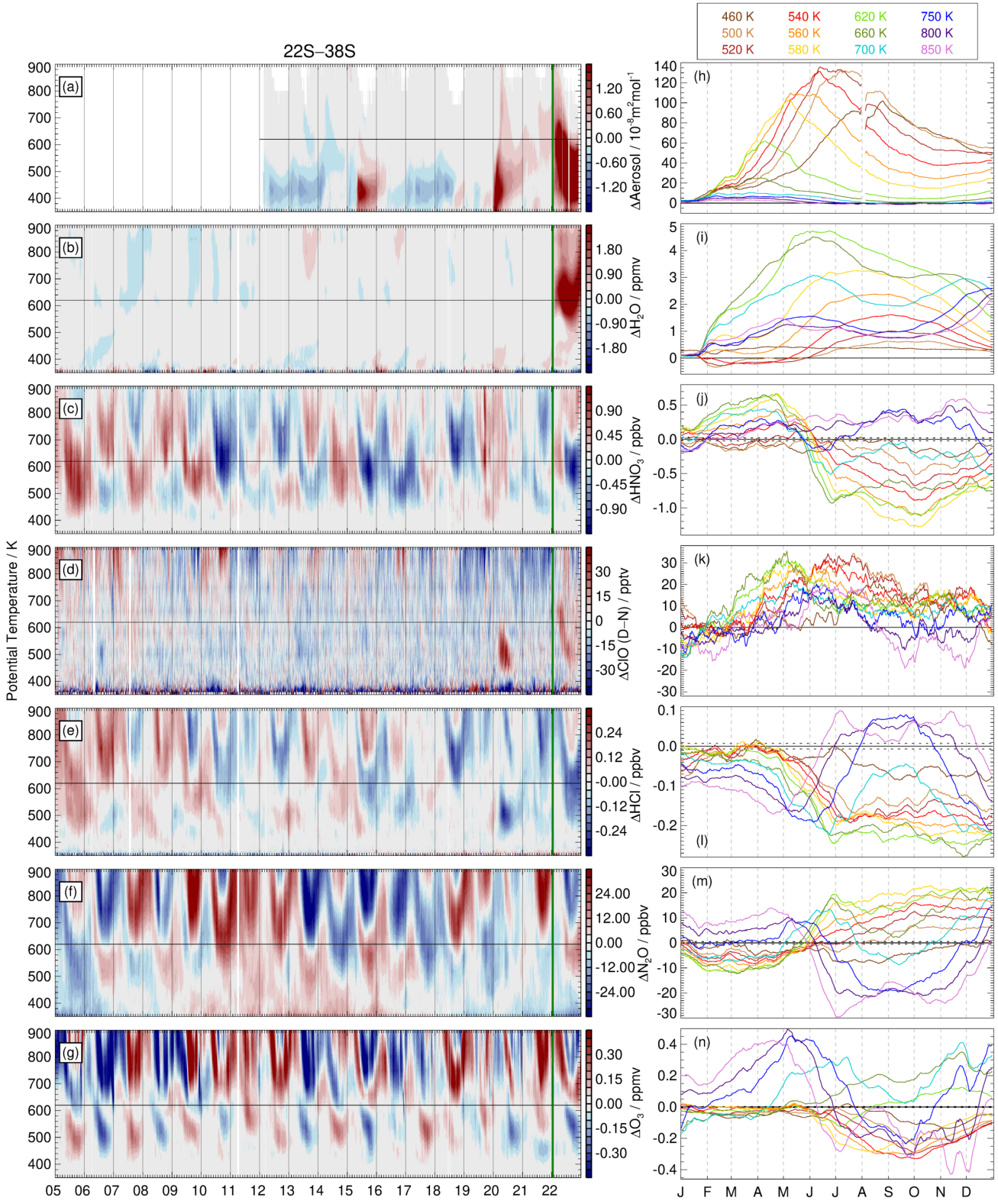

Figure S3. As Figure 3 in the main text, but for the 22°S–38°S EqL band.

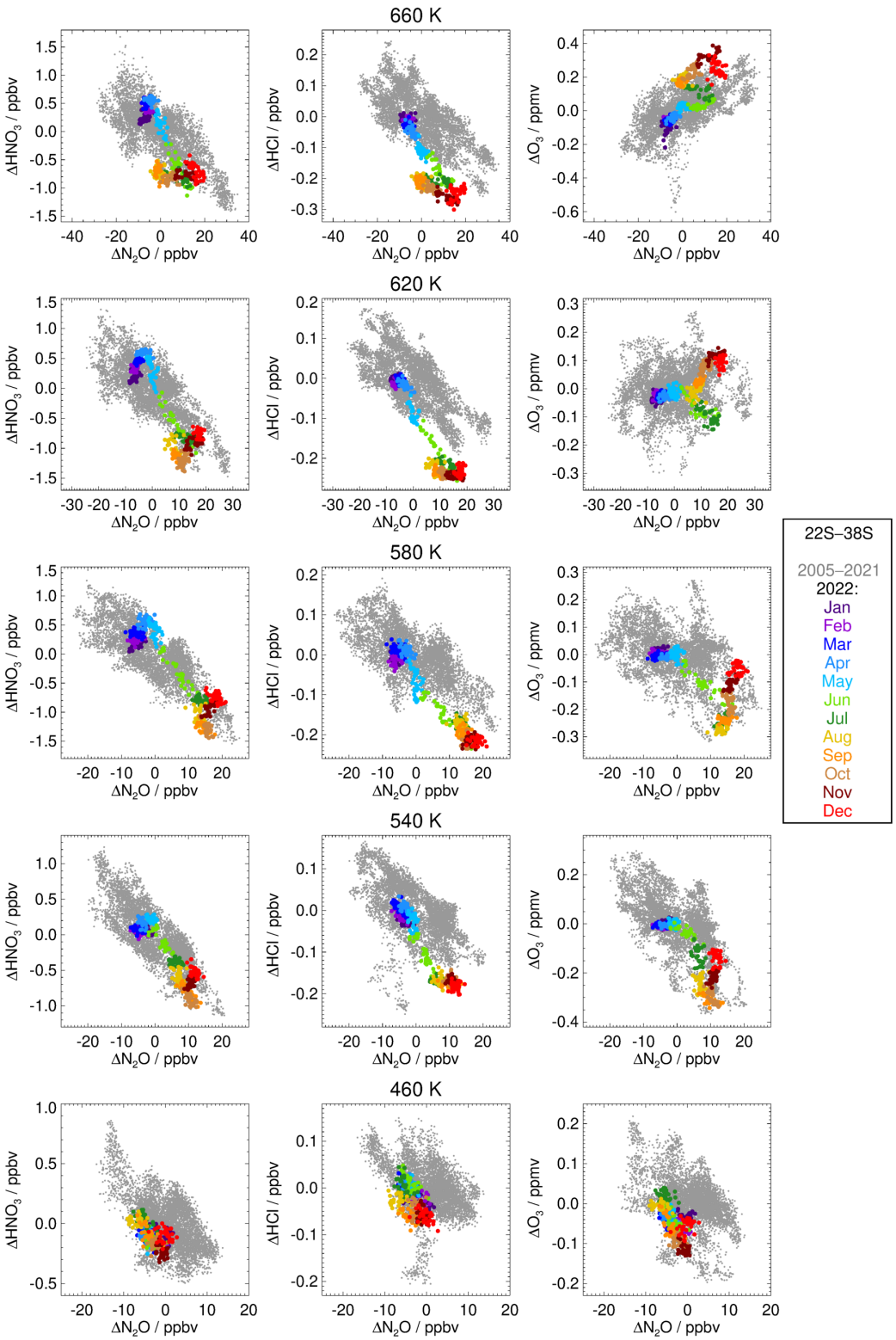

Figure S4. As Figure 4 in the main text, but for the 22°S–38°S EqL band.

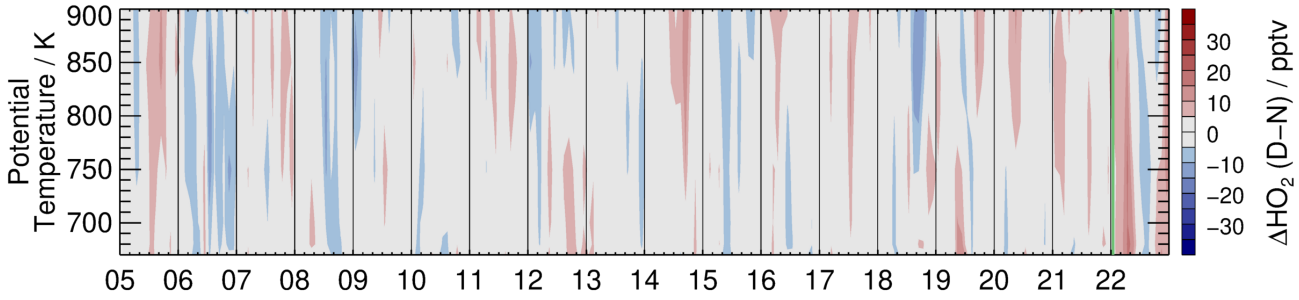

Figure S5. Potential temperature / time section, as in Figures 3 and 8 in the main text, but for the anomaly in MLS monthly HO_2 (day minus night) in the 6°S–22°S EqL band.