

Supplementary Materials

2 Supplementary Figures

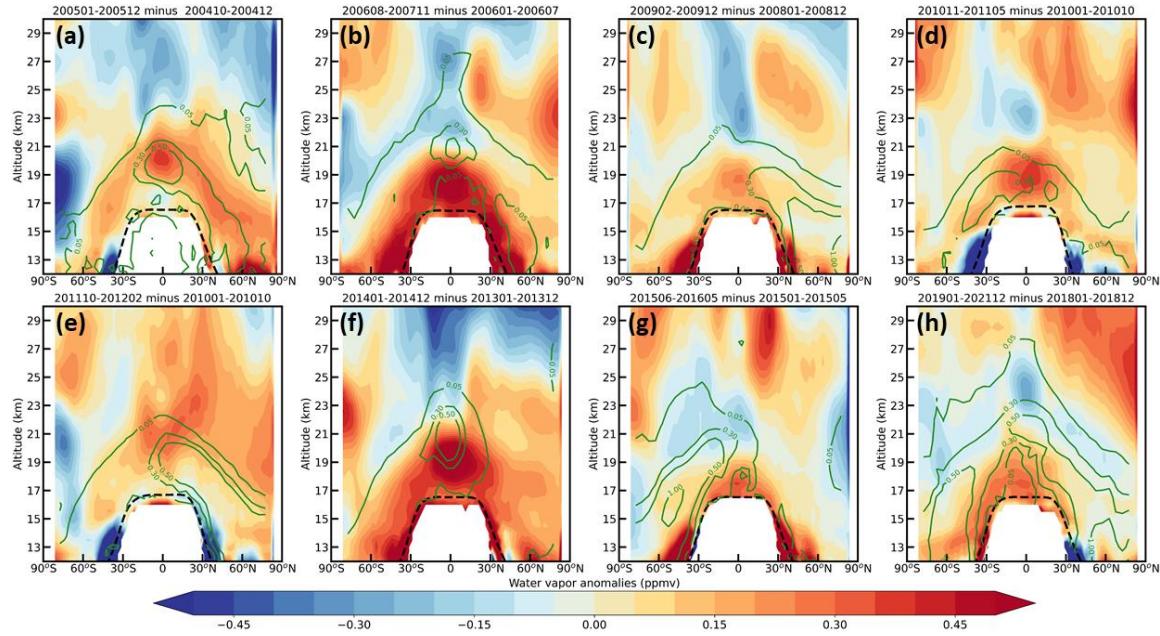


Figure S1. Observed averaged SWV anomalies relative to climatology (2005–2021) during periods of enhanced LS AOD are denoted by the shading. The green lines denote the aerosol extinction anomalies relative to the climatology of 2000–2005 from GloSSAC. (a) Difference between the mean SWV anomalies during January–December 2005 and those during October–December 2004. (b) Difference between the mean SWV anomalies during August 2006 – November 2007 and those during January–July 2006. (c) Difference between the mean SWV anomalies during February–December 2009 and those during January–December 2008. (d) Difference between the mean SWV anomalies during November 2010–May 2011 and those during January–October 2010. (e) Difference between the mean SWV anomalies during November 2011–February 2012 and those during January–October 2010. (f) Difference between the mean SWV anomalies during January–December 2014 and those during January–December 2013. (g) Difference between the mean SWV anomalies during June 2015–May 2016 and those during January–May 2015. (h) Difference between the mean SWV anomalies during January 2019–December 2021 and those during January–December 2018.

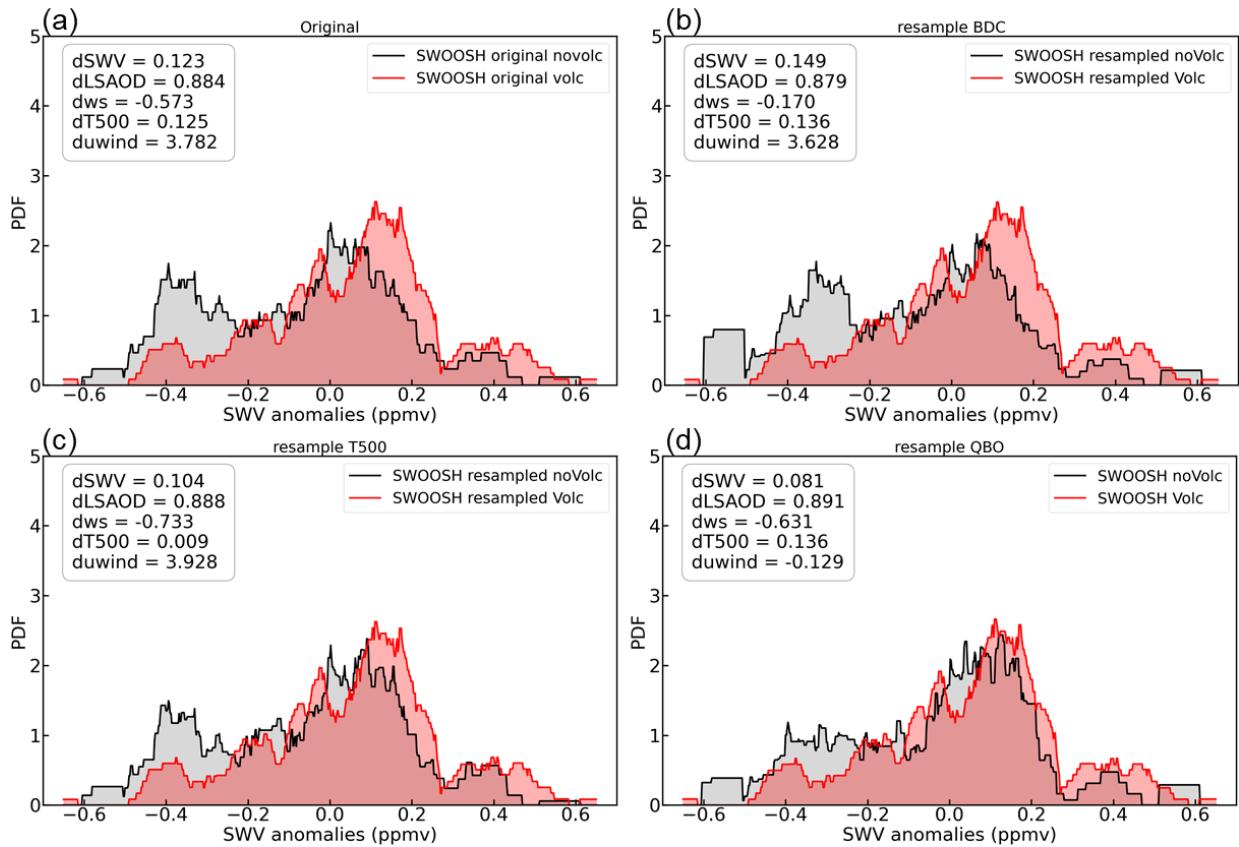


Figure S2. Probability density functions (PDFs) of tropical (30° N– 30° S) SWV anomalies at 82 mb for volcanic/wildfire and non-volcanic/wildfire months. (a) Original dataset; (b–d) after resampling to control for variability associated with (b) BDC, (c) T500 (representing both ENSO and global warming), and (d) QBO. Numbers in the upper-left corner of each panel denote the differences between volcanic/wildfire and non-volcanic/wildfire months in SWV (dSWV), LS AOD (dLS AOD), BDC (dws), T500 (dT500), and QBO (duwnd). Red and black lines represent PDFs for volcanic/wildfire and non-volcanic/wildfire months, respectively.

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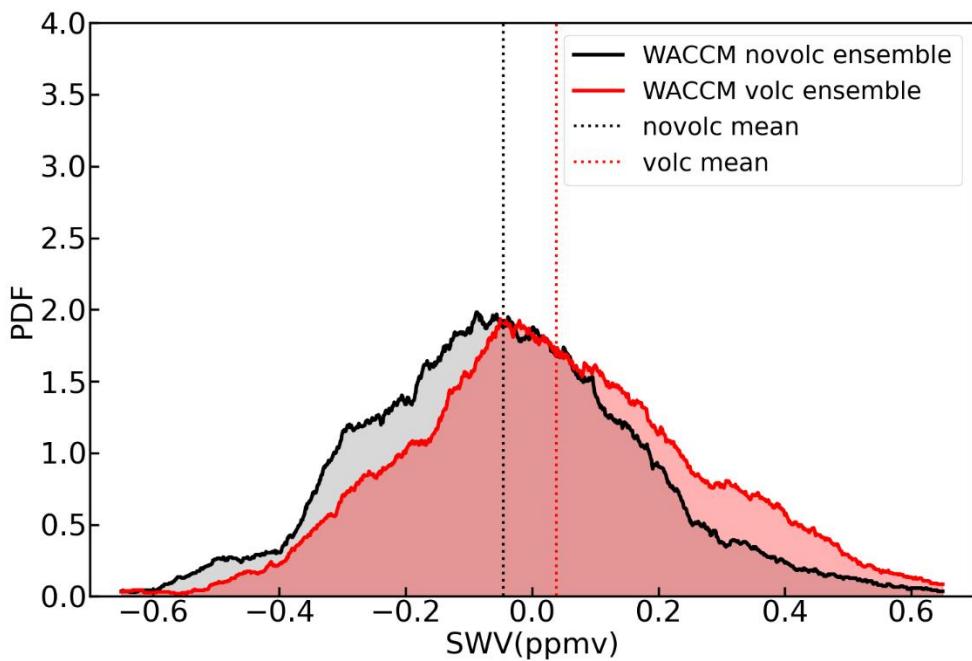
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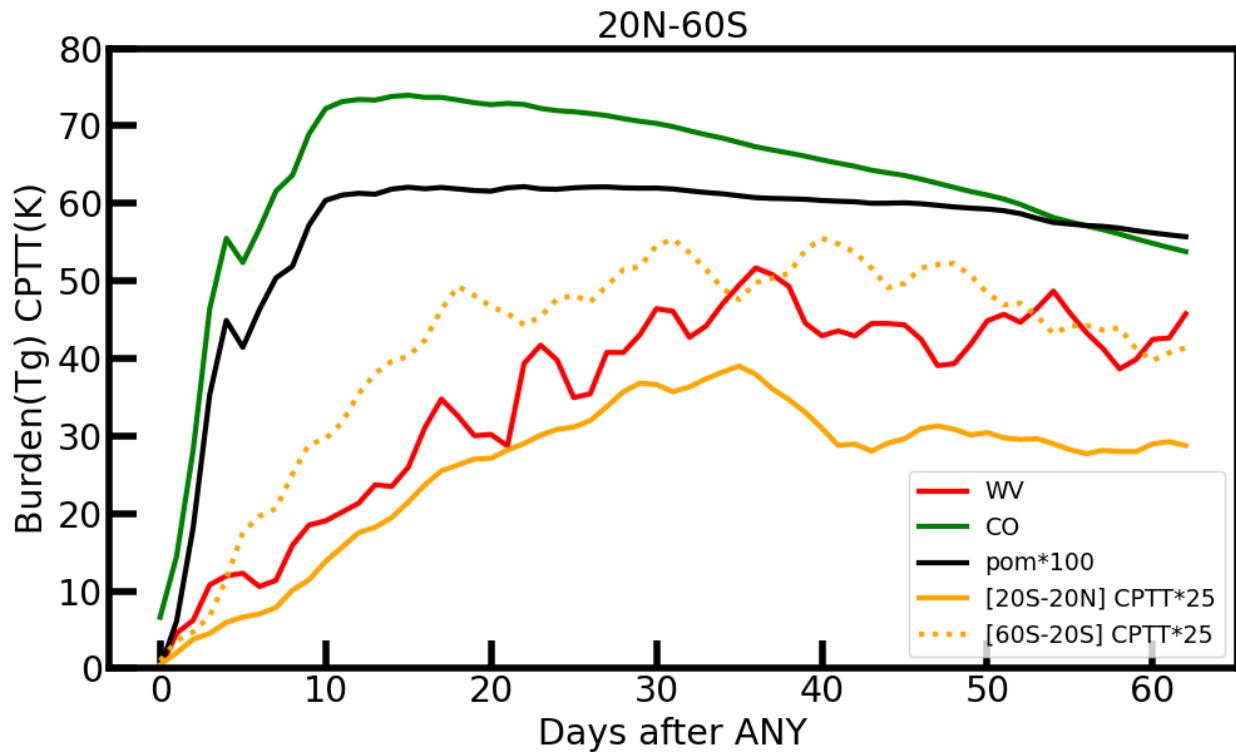
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Figure S3. Model-simulated SWV anomaly PDFs for volcanic/wildfire and non-volcanic/wildfire periods, derived from member-level PDFs averaged across the ensemble. Mean values of volcanic/wildfire and non-volcanic/wildfire periods are denoted by red and black dashed lines, respectively.



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35 **Figure S4.** Simulated changes in stratospheric constituents and cold point tropopause
 36 temperature (CPTT) associated with the 2020 Australian wildfire (ANY) over the tropics and
 37 Southern Hemisphere midlatitudes (20° N– 60° S). Red and green lines denote the stratospheric
 38 burdens of water vapor (SWV) and carbon monoxide (CO), respectively. The black line shows
 39 the stratospheric burden of organic carbon (OC) aerosols multiplied by 100 for clarity. The
 40 orange line represents CPTT anomalies (relative to the non-ANY simulations) multiplied by 25.
 41 Solid and dashed orange lines indicate CPTT anomalies averaged over the tropics (25° N– 25° S)
 42 and Southern Hemisphere midlatitudes (60° S– 20° S), respectively.

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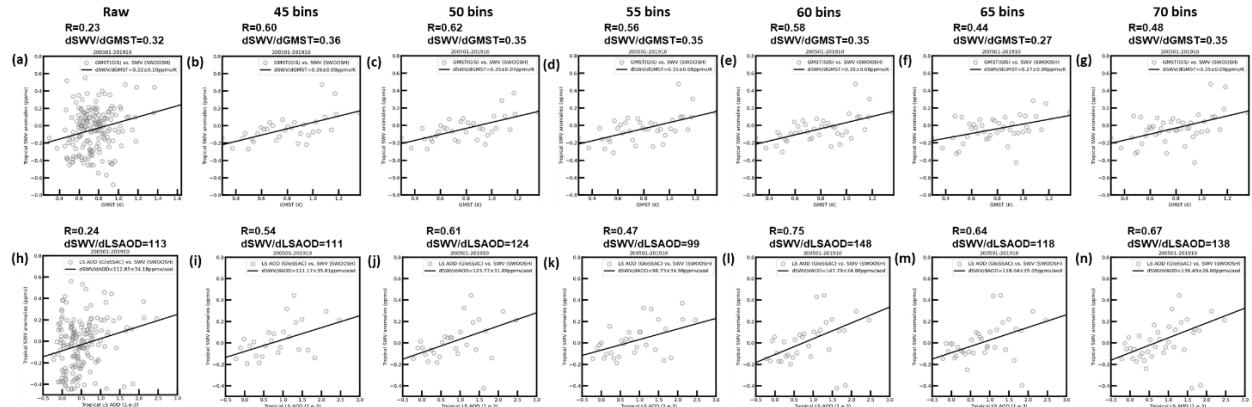


Figure S5. Relationships between stratospheric water vapor (SWV) and its driving factors derived from observational data during January 2005–October 2019. (a-g) Relationships between SWV and global mean surface temperature (GMST). (a) The regression based on raw observational data. (b-g) The same relationship but with GMST uniformly divided into 45, 50, 55, 60, 65, and 70 bins between its minimum and maximum values, and mean SWV anomalies in each bin are used for regression. (h-n) Same as (a-g), but for the relationships between SWV and lower stratospheric aerosol optical depth (LS AOD).