Supplementary material for

Two-years of stratospheric chemistry perturbations from the 2019/2020 Australian wildfire smoke

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Figure S1. Analysis of homogeneous mixed aerosol assumption for HCl solubility in CESM1-CARMA. Total sulfate mass mixing ratios in mixed sulfate (a), pure sulfate (b). Total organics including primary and secondary organics (c), sulfate fraction in mixed aerosols assuming all organics are in mixed aerosol bins for HCl solubility linearization

20 for both control and wildfire simulations. Results for HCl absolute values (e) and anomalies (f) for homogeneous and LLPS mixed aerosol assumptions of HCl solubility. Offsets of model absolute values compared to observations in (e) (-0.16 ppb) have been normalized to the MLS climatology January 1 value.



Sulfate and organic mass fractions

25 Figure S2. CESM1-CARMA mass fraction of background sulfate and organic aerosols over 55–40°S. background SOA values are approximately 4 times larger than Murphy et al., (2021). See main text.



Figure S3. Comparison of OMPS-LP and CESM1-CARMA 675 nm aerosol extinction coefficient for southern midlatitudes (a, and b) and polar regions (c, and d). Values show ratio of 2020–2021 values to background levels. The OMPS-LP background is from volcanically clean years. i.e. 2012, 2013, 2014, and 2017. The CESM1-CARMA background is the control run. Vertical dotted line in c, and d represents the date that 10 hPa zonal wind transition from westerly winds to easterly winds in ERA5.



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Figure S4. Analysis of seasonality of ANY organic induced seasonal partitioning. (a) 2020 CESM1-CARMA ClONO₂ and HCl (with inverted y axis) for the wildfire and LLPS control (1/4 SOA) cases that shows majority of Cl from HCl partitioned into ClONO₂ with a peak in austral winter. (b) 2020 CESM1-CARMA Cl/ClO for the control and the wildfire run highlighting the seasonality of active Cl partitioning (Cl is more likely to be in ClO in austral winter). Box model NO₂/NO for austral summer (c) and winter (d). CESM1-CARMA 3 pm values of NO₂ and NO

over 2020 for the wildfire case (e). CLO + NO/ClO + NO₂ in a box model (<u>https://github.com/KaneStone/BushfireChemModel/releases/Hunga_v1.0</u>) and CESM1-CARMA wildfire case (f).

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Figure S5. Absolute polar HCl and ClONO₂ values for 100 hPa (a,b) and 68 hPa (c,d) highlighting the discrepancy between the CESM1-CARMA control HCl loss compared to MLS and agreement between the CESM-CARMA wildfire case in 2021.