

1 *Supplement of*
2 ***In situ* measurements of perturbations to stratospheric aerosol**
3 **and modeled ozone and radiative impacts following the 2021 La**
4 **Soufrière eruption**

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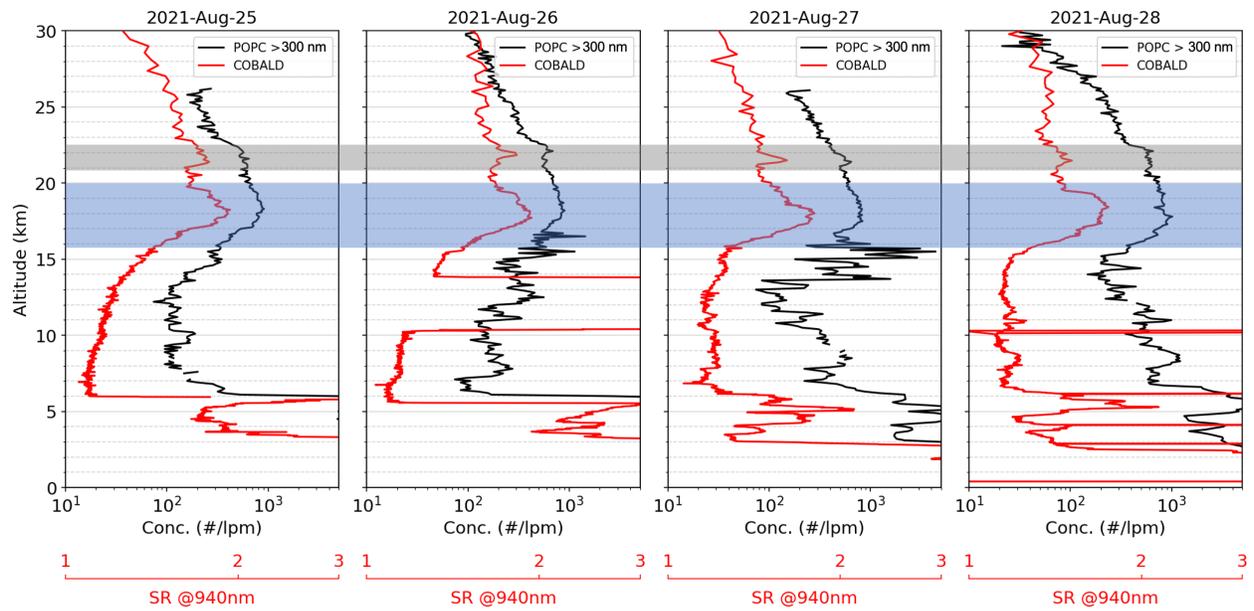
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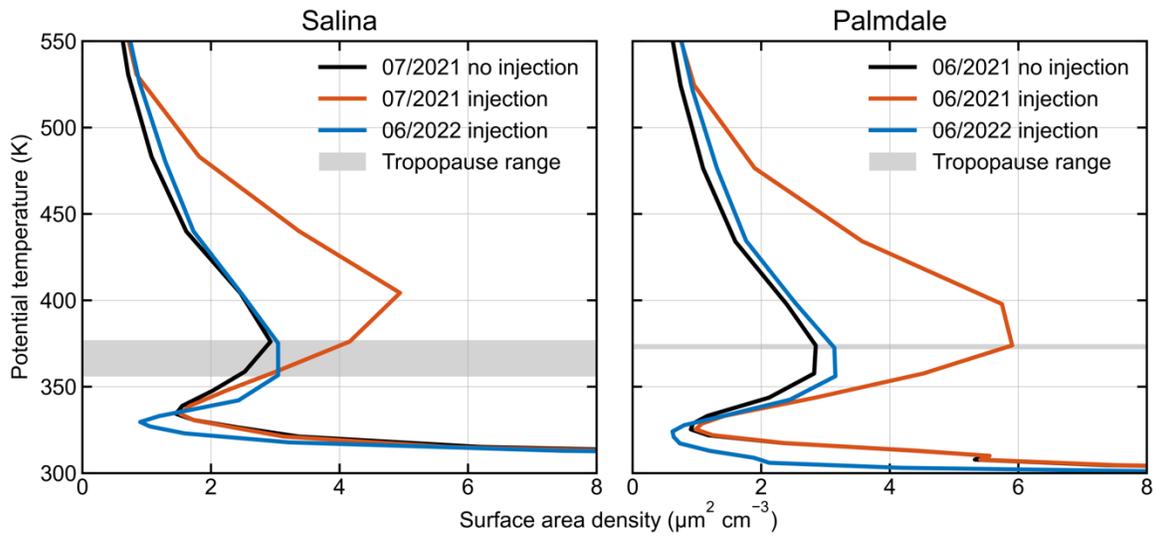
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30 Figures S1 to S10



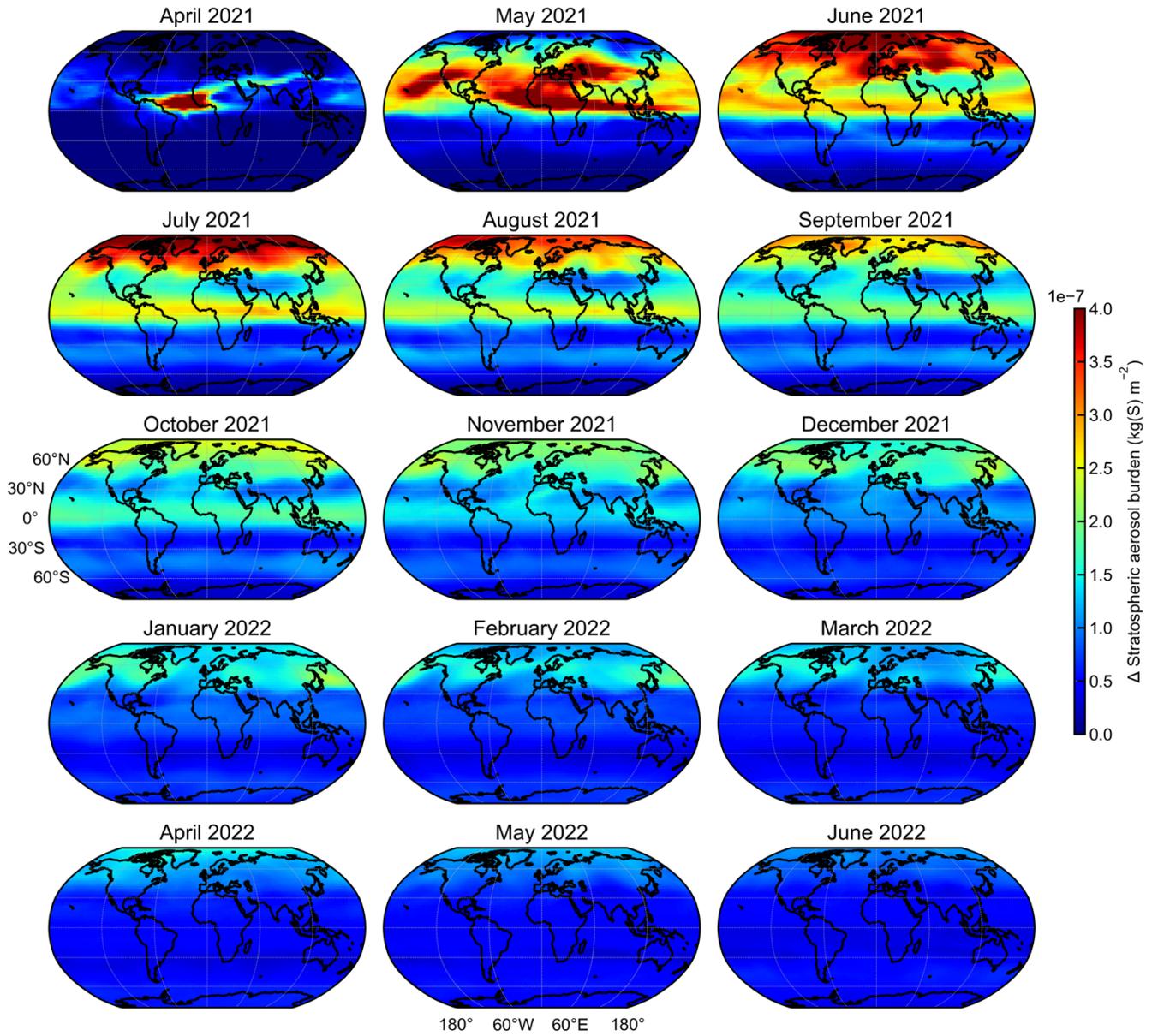
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Figure S1. Aerosol vertical profiles from nighttime DCOTSS balloon-borne observations using POPC and COBALD instruments over Salina, Kansas in August 2021. Scattering ratios (SR) at 940 nm were calculated from COBALD backscatter measurements. The blue shaded area shows aerosol enhancements from the lower volcanic plume, while the grey shaded area shows the influence of the transient filament excursion of the tropical upper plume.



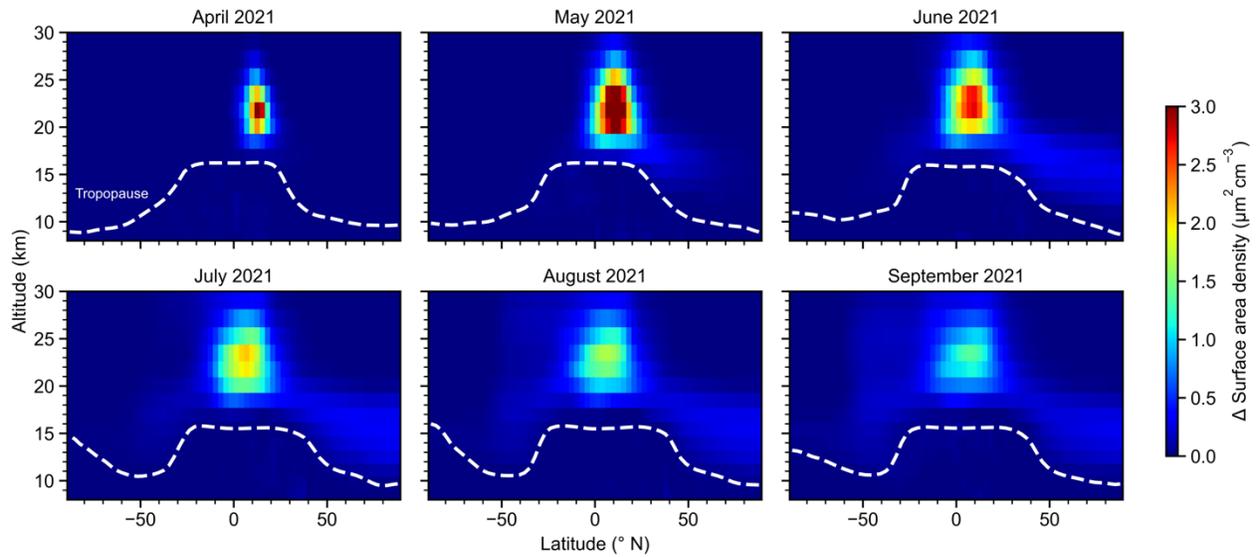
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Figure S2. SOCOL-AERv2 surface area density vertical profiles. The black line is the no-injection simulation. The orange and blue lines are model runs with a 0.4 Tg SO₂ injection. The percent difference between the 2022 injection case (blue line) and the 2021 no-injection case (black line) averaged over the range of 375-530 K is 12% and 16% for Salina and Palmdale, respectively.

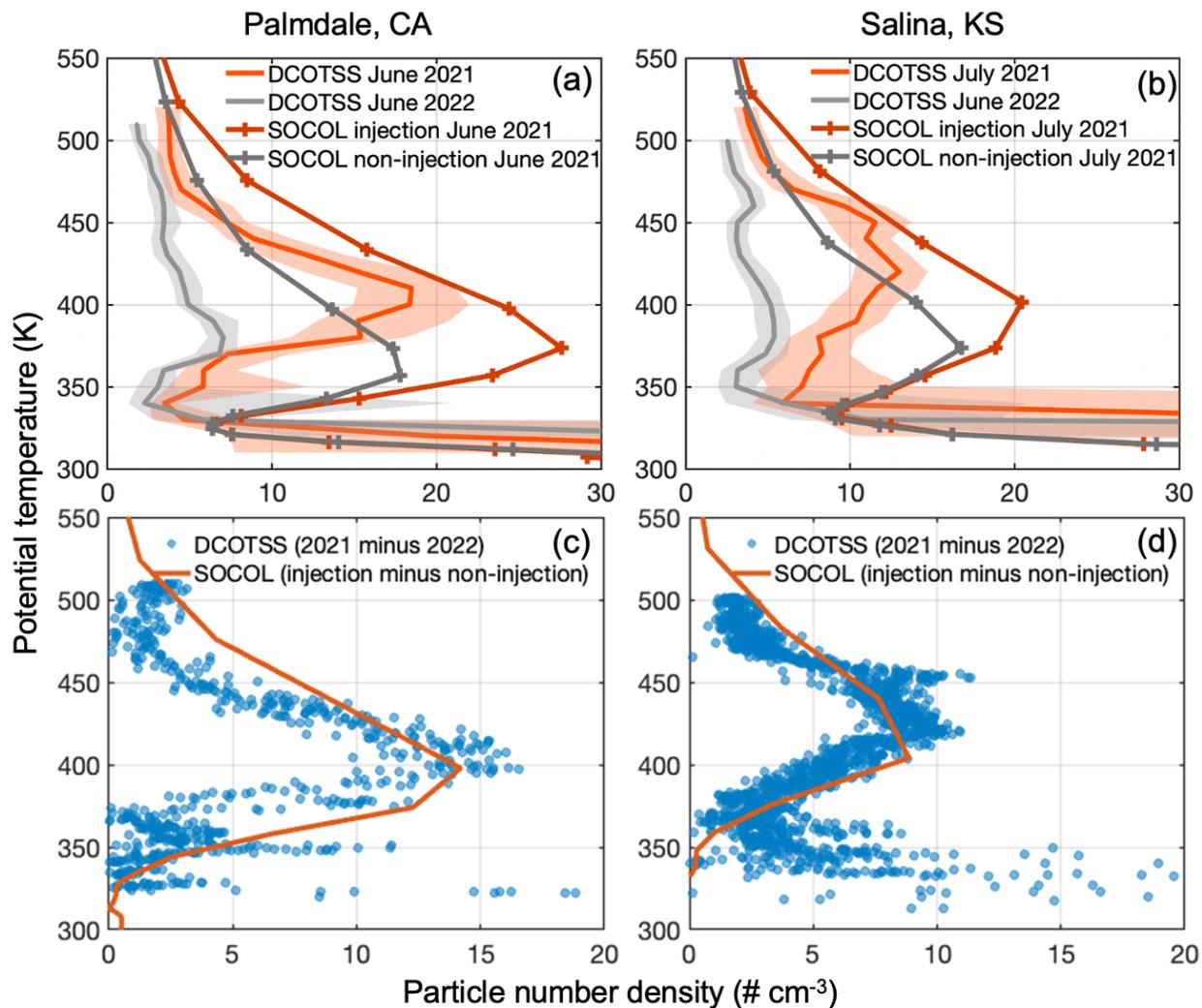


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Figure S3. Global map of the stratospheric aerosol burden from the La Soufrière eruption. The aerosol burden is plotted as the difference between the 0.4 Tg injection and no-injection SOCOL-AERv2 simulations.

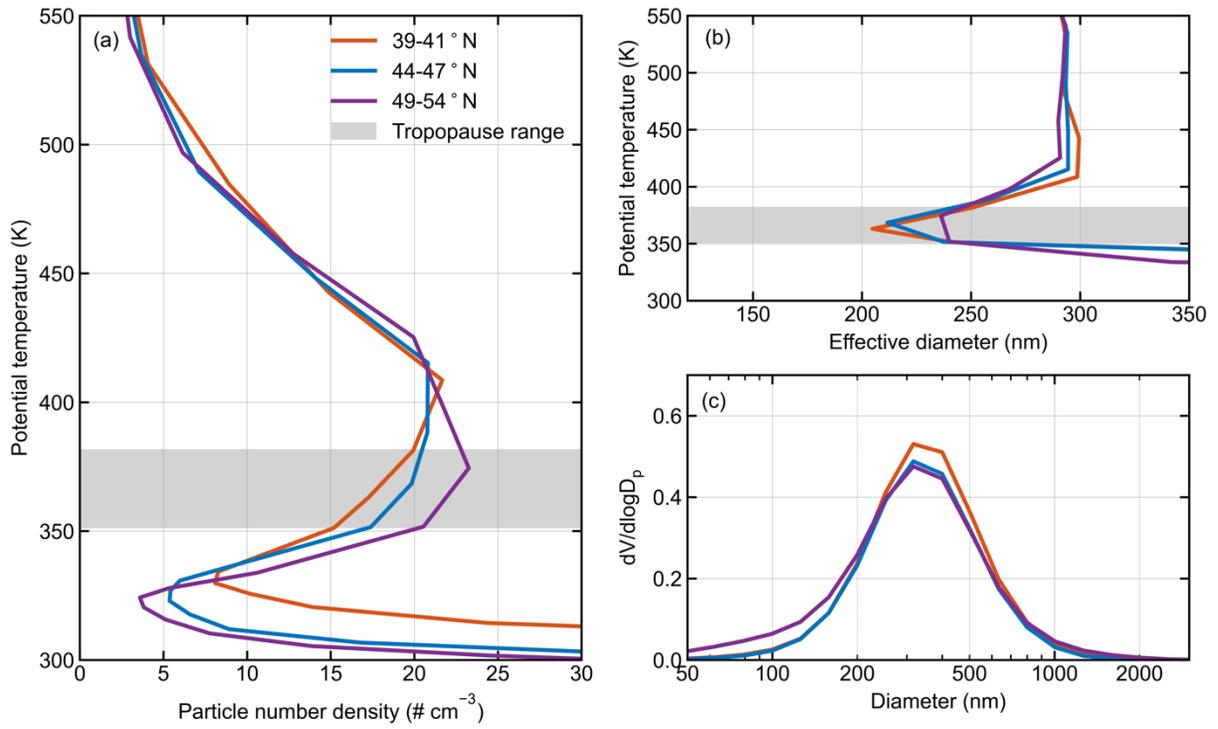


44 **Figure S4. Analogous to Figure 4 in the main text but for the upper plume simulation. Latitude and altitude distributions**
 45 **of aerosol surface area density (SAD) difference between injection and non-injection simulations performed by SOCOL-**
 46 **AERv2 from April to September 2021. Delta SAD was averaged zonally and monthly and used all aerosol sizes in the**
 47 **calculation. The white dashed lines indicate the WMO-defined tropopause. The upper plume was simulated by injecting**
 48 **0.1 Tg of SO₂ in one grid box at 42 hPa (~22 km).**



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Figure S5. Comparisons of particle number density profiles between SOCOL-AERv2 model simulations and DCOTSS aircraft measurements. (a) and (c) present vertical profiles over Palmdale (averaged over 32-38° N, 115-121° W), while (b) and (d) present vertical profiles over Salina (averaged over 36-42° N, 95-101° W). The absolute number density profiles are presented in (a) and (b), while the number density enhancement profiles are presented in (c) and (d). Shaded areas in (a) and (b) correspond the 10th and 90th percentiles of the DCOTSS aircraft measurements.



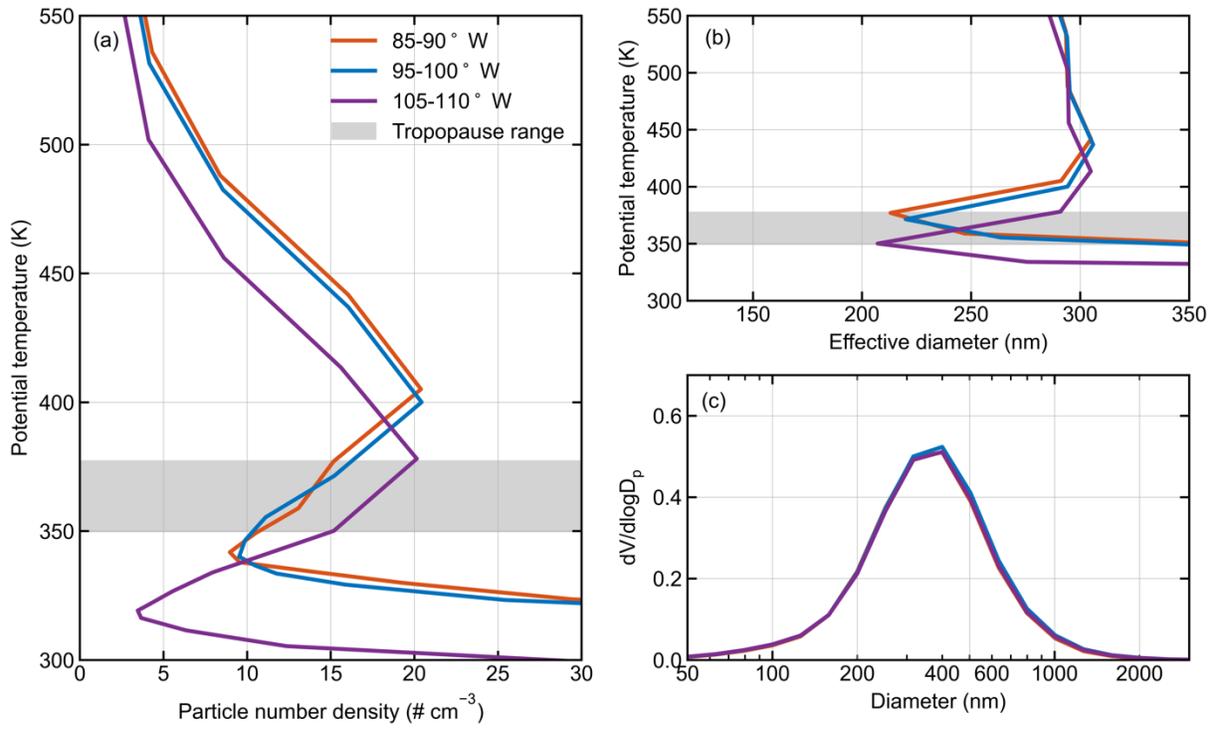
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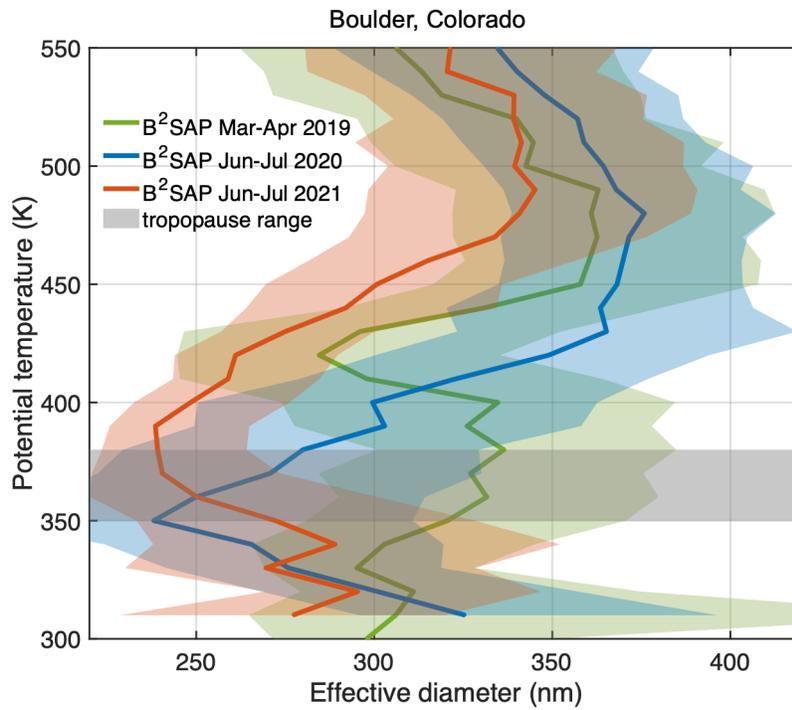
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Figure S6. Model equivalent to Figure 6. Panel (c) is the size distribution at the plume peak. The peaks of 33-37° N, 44-47° N, and 49-54° N are at pressure levels 89 hPa, 89 hPa, and 143 hPa, respectively, which correspond to a potential temperature of 409, 415, and 374, respectively. The grey tropopause range in (a) and (b) is the modeled tropopause range.



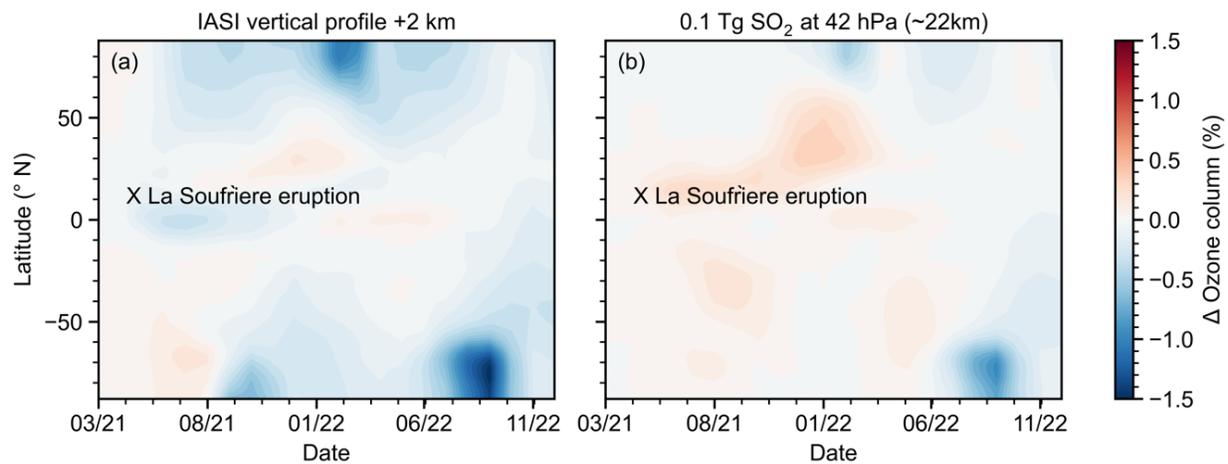
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60 **Figure S7. Model equivalent to Figure 7. Panel (c) is the size distribution at the plume peak. The peaks of 85-90° W, 95-100°**
 61 **W, and 105-110° W are all at pressure level 89 hPa which correspond to potential temperatures of 405, 400, and 378,**
 62 **respectively. The downward shift of the 105-110 W profile in panels (a) and (b) is an artifact from the calculation of potential**
 63 **temperature. This feature is not present with pressure as the vertical coordinate. The grey tropopause range in (a) and (b)**
 64 **is the modeled tropopause range.**

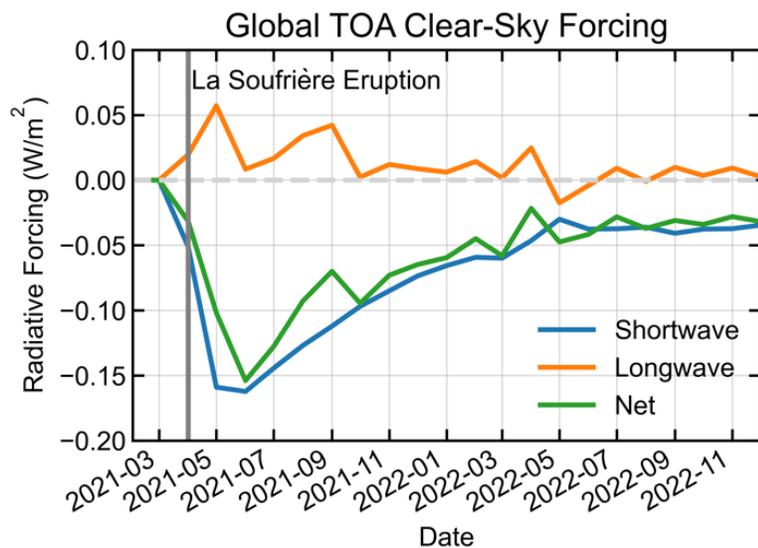


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Figure S8. Vertical profiles of aerosol effective diameter over Boulder, Colorado, during March-April 2019, June-July 2020, and June-July 2021 from balloon-borne measurements as part of the Baseline Balloon Stratospheric Aerosol Profiles (B²SAP) project (Todd et al., 2023). Data from March-April 2019 was selected to represent unperturbed condition before the 2019 Raikoke eruption.



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Figure S9. Modeled zonal-averaged ozone column change for two sensitivity simulations: (a) a 2 km upward shift of the IASI SO₂ vertical profile and (b) 0.1 Tg injected at 42 hPa (~22 km) in a single model grid cell above La Soufrière. Ozone column change is shown as the percent difference between the SO₂ injection simulation and the no-injection simulation.



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 75 **Figure S10. Global top-of-atmosphere (TOA) clear-sky radiative forcing following the La Soufrière eruption based on**
 76 **SOCOL-AERv2 simulations. Radiative forcing is calculated as the difference between the SO₂ injection simulation and the**
 77 **no-injection simulation.**

78 **References**

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 80 Stratospheric Aerosol Profiles (B2SAP)—Systematic Measurements of Aerosol Number
 81 Density and Size. *Journal of Geophysical Research: Atmospheres*, 128(12),
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