How do aerosol modeling assumptions and parameterizations affect stratospheric sulfate aerosol following the 1991 Pinatubo volcanic eruption?

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I. Motivation

- A variety of assumptions in aerosol treatment can lead to very different microphysical evolution, development, and direct radiative impacts
- Using a modified version of the Energy Exascale Earth System Model (E3SMv2) to improve representation of stratospheric sulfate formation/evolution from the 1991 Pinatubo eruption for the CLimate impact: Determining Etiology thRough pAthways (CLDERA) project (*Brown et al., 2024*).

II. Model and Observational Datasets

- Model simulations
  - **E3SMv2-PA, E3SMv2-SPA, E3SMv2-presc**
    - Prognostic default model **(PA),** prognostic stratospheric sulfate aerosol (**SPA**), and prescribed stratospheric aerosol forcing (**presc**)
  - Community Earth System Model with the Whole Atmosphere Chemistry Climate Model (**CESM2-WACCM**) (*Mills et al., 2016, 2017*)
    - Full-chemistry and prognostic stratospheric sulfate aerosol
- Observational datasets
  - Stratospheric mass burden – High Altitude Infrared Radiation Sounder (**HIRS**) (*Baran and Foot, 1994*) and **SAGE-3λ** *(Revell et al., 2017)*
  - Size distributions and effective radius - Wyoming optical particle counter (**WOPC**) (*Deshler et al., 1993*)

III. Modal aerosol distribution assumptions

- Default E3SMv2 (E3SMv2-presc) prescribes stratospheric sulfate from explosive volcanic eruptions. When configured to simulate sulfate formation and evolution (E3SMv2-PA), sulfate lifetime is too short (Fig. 4).
- Improved model referred to as E3SMv2-SPA (Prognostic Stratospheric sulfate Aerosol). Major modifications to the Modal Aerosol Module (*Liu et al., 2012, 2016*) are shown in Fig. 2; the impacts of these modifications on stratospheric aerosol size distributions are shown in Fig. 3.

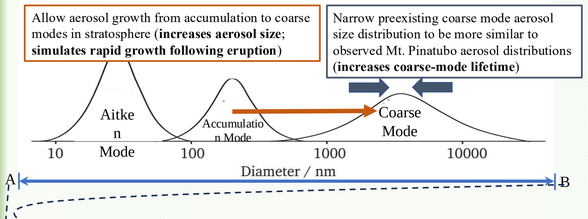

Figure 2: Major changes made to E3SMv2-PA aerosol size modes in order to prognostically calculate stratospheric sulfate formed from the volcanic emission of sulfur dioxide in the stratosphere.

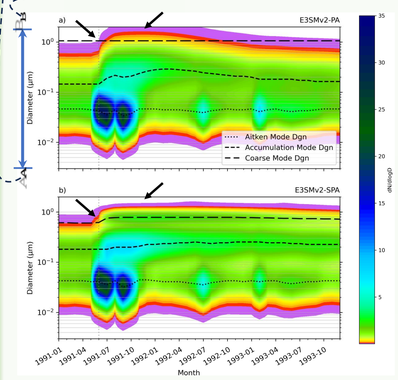

Figure 3: Globally averaged stratospheric lognormal aerosol size distributions from 1991–1993 for E3SMv2-PA, E3SMv2-SPA, and CESM2-WACCM. The dotted, dashed, and long dash lines indicate geometric mean diameters (D_{gn}) of the Aitken, accumulation, and coarse aerosol modes, respectively. The color contour is lognormally scaled. The Pinatubo eruption is marked with the dotted gray line at June 1991. The arrows indicate the impacts of the major microphysical modifications. (**left arrow**) the formation of a coarse mode in E3SMv2-SPA, denoted by an increase in Coarse mode D_{gn}; (**right arrow**) a wide accumulation mode distribution in E3SMv2-PA (fast removal), replaced by a narrower coarse mode distribution which better matches observations of post-Pinatubo stratospheric aerosol size distributions.

A diverse set of aerosol assumptions including aerosol modal characteristics, atmospheric chemistry, nucleation scheme, and injection height all contribute to accurately modeling sulfate aerosol formation and lifetime following Mt. Pinatubo. The best models in our study simulate reasonably well compared to global remote sensing, but regional sampling over Laramie, WY indicates an underestimation in aerosol size that could lead to overestimation in modeled aerosol shortwave scattering.

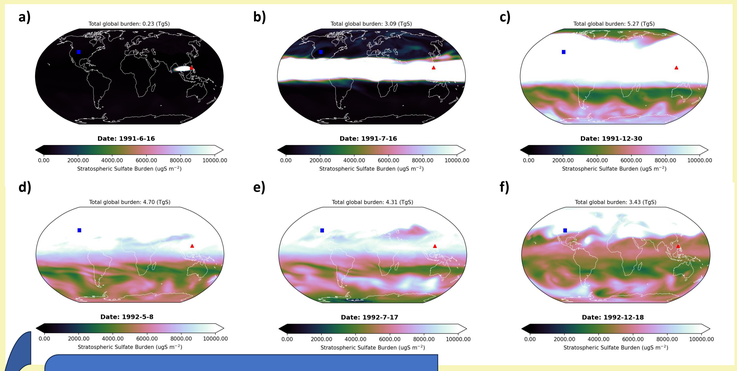

Figure 1: Daily, stratospheric sulfur burden for ~18 months following the Mt. Pinatubo eruption on June 6th, 1991 (a). The red triangle marks Mt. Pinatubo, and the blue square marks our sampling region of Laramie, WY. Southern Hemisphere aerosol appearing in (c) is due to the Cerro Hudson in southern Chile that occurred on August 8th, 1991. Peak aerosol size over Laramie occurs in May 1992 (d) (Fig. 6d).

Total Mass
Gradual removal and spread to the poles

Aerosol size distributions
~11 months post eruption, models fail to simulate peak aerosol size

Aerosol scattering
Overestimation in model scattering at wavelength of peak insolation by models due to size underestimation

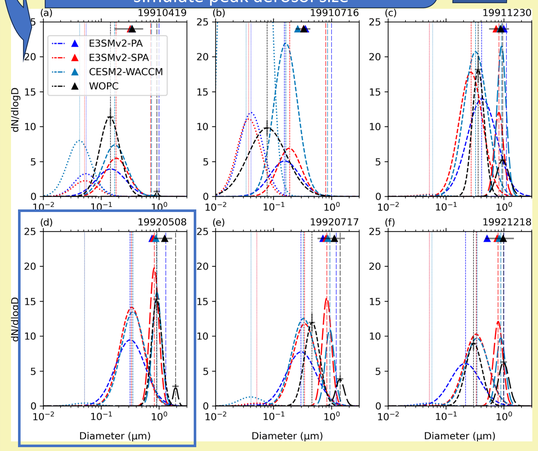

Figure 6: Stratospheric aerosol size distributions from models and WOPC (black) corresponding to stratospheric burden in Fig. 1. WOPC samples are taken from the 18 km measurements and matched to the nearest model height and grid cell over Laramie, Wyoming (41.3° N, 105° W). The dotted, dashed, and long-dash lines indicate geometric mean diameters (vertical) and dN/dlogD size modes (curves) of the Aitken, accumulation, and coarse aerosol modes, respectively. Triangles denote effective diameters (D_{eff}) derived from the size distributions. Uncertainties in the WOPC diameter, number, and D_{eff} are denoted by gray bars at the peak of the distributions and on the markers.

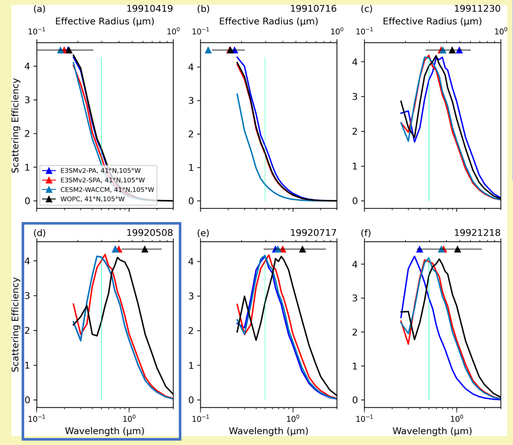

Figure 7: Daily 18 km scattering efficiency (Qs) using the effective size parameter ($x_{eff} = 2\pi R_{eff}/\lambda$) and sulfate refractive index at 0% relative humidity (Hess et al., 1998). Effective radii (R_{eff}) from models and WOPC are calculated at 18 km and are marked by colored triangles at the top of the plot. The bottom x axis is wavelength (; 0.1–3 μm). The top axis is Reff (0.1–1 μm) used in the calculation of Qs via xeff. The vertical turquoise line marks the solar blackbody wavelength of maximum irradiance (0.5 μm).

References:
- Baran and Foot, 1994 (10.1029/94JD02044)
- Brown et al., 2024 (10.5194/gmd-17-5087-2024)
- Deshler et al., 1995 (10.1029/93GL01337)
- Liu et al. (2012) (10.5194/gmd-5-709-2012)
- Liu et al. (2016) (10.5194/gmd-9-505-2016)
- McCormick and Veiga, 1995 (10.1029/91GL02790)
- Mills et al. (2016) (10.1002/2015JD024290)
- Mills et al. (2017) (10.1002/2017JD027006)
- Quaglia et al., 2023 (10.5194/acp-23-921-2023)
- Revell et al. (10.5194/acp-17-13139-2017)
- Stenchikov et al., 2021 (10.1029/2020JD033829)

See QR code for Brown et al. (2024) from which this poster was created

IV. Full Chemistry vs. Simplified Chemistry

- Models simulate the formation of sulfate aerosol (H_2SO_4) based on emitted sulfur dioxide (SO_2) and atmospheric hydroxyl radical (OH) concentrations:

$$SO_2(gas) + OH \rightarrow H_2SO_4(aerosol)$$

- E3SM assumes a prescribed atmospheric OH concentration (non-limiting), whereas the full-chemistry CESM2-WACCM represents the consumption of OH via this reaction, limiting the formation of H_2SO_4 in CESM2-WACCM.
- Leads to: **(1)** earlier formation of sulfate in the stratosphere in E3SM (A; Fig. 4) and lower max aerosol burden in the stratosphere for CESM2-WACCM (B; Fig. 4); **(2)** fewer but larger aerosol in CESM-WACCM due to growth of fewer initial nucleated particles (not shown)

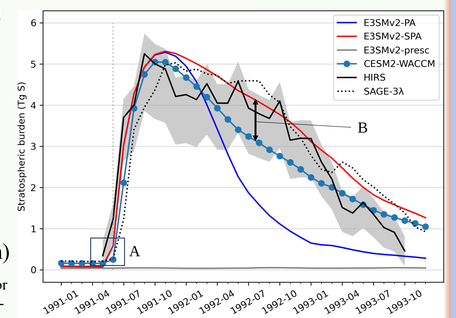

Figure 4: Stratospheric sulfate burden (TgS) for model simulations, as well as HIRS and SAGE-3λ remote sensing observations.

III. Plume height

- Under Pinatubo stratospheric conditions, models that form sulfate **<~20 km tend to transport aerosol north** (**shorter lifetime; larger D_{gn}**) while **sulfate >~20 km tends to be confined to the Tropics** (**longer lifetime; smaller D_{gn}**) (*Quaglia et al., 2023; McCormick and Veiga, 1995*)
- Volcanic ash and larger particles can also increase plume lifetime/ by heating/lofting the plume through plume radiation absorption (*Stenchikov et al., 2021*).

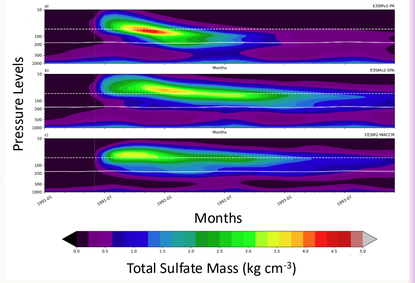

Figure 4: : Vertical distribution of global mean sulfate aerosol mass concentration from 1991-1993 for (a) E3SMv2-PA, (b) E3SMv2-SPA, and (c) CESM2-WACCM. The white line denotes the model tropopause, the dashed white line denotes the nearest model level to the 20 km geopotential height. E3SMv2-SPA tended to be confined more to the Tropics and had slightly longer lifetime than CESM2-WACCM

VI. Aerosol Stratospheric Effective Radius

- Effective radius (R_{eff}) is the area-weighted aerosol radius and is a good indicator of a size distribution's optically relevant size.
- All models underestimate R_{eff} compared to WOPC atmospheric profiles due to models not simulating large enough coarse mode aerosol (Fig. 6).
- Largest size differences occur ~12 months post-eruption
- This may be related to nucleation scheme, atmospheric chemistry treatment, injection height, and/or missing interparticle interactions (i.e., van der Waal forces)

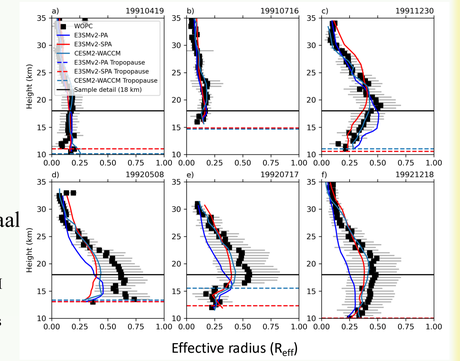

Figure 5: Vertical profiles of daily WOPC R_{eff} over Laramie (41°N 105°W) compared to E3SM and CESM2-WACCM. Dashed lines show the model tropopause and the solid black line marks the 18 km sample level corresponding to size distributions in Fig. 6.