Radiative forcing by super-volcano supervolcano eruptions

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Key Points:

- The linear RF_ERF dependence on AOD breaks down for eruptions larger than

 Mt. Pinatubo
- The RF-ERF to AOD ratio has a time-after-eruption dependence on eruption latitude
- Temperature and RF_ERF peak values has a linear dependence and reaches an upper limit

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Abstract

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We investigate the climatic effects of volcanic eruptions spanning from Mt. Pinatubo-13 sized events to reviewer1: [L11] supercruptions. The study is based on ensemble simulations 14 in the Community Earth System Model Version 2 (CESM2) climate model using the Whole 15 Atmosphere Community Climate Model Version 6 (WACCM6) atmosphere model. Our 16 analysis focuses on the impact of different SO₂-amount injections on stratospheric aerosol 17 optical depth (AOD), reviewer1: [L15] effective radiative forcing (ERF), and global temperature 18 mean surface temperature (GMST) anomalies. Unlike the traditional linear models model 19 used for smaller eruptions, our results reveal a non-linear relationship between RF-is typically 20 found between ERF and AOD for larger eruptions. We also further uncover a notable 21 time-dependent decrease in aerosol forcing efficiency (ERF normalised by AOD) across 22 all eruption magnitudes during the first post-eruption year. In addition, the study re-23 veals that larger the largest as compared to medium-sized Pinatubo-sized eruption events 24 produce a delayed and sharper peak in AOD, and a longer-lasting temperature CMST 25 response while the time evolution of RF-ERF remains similar between the two eruption 26 types. We find that the peak ERF approaches a limiting value, and that the peak GMST 27 response follows linearly, effectively bounding the GMST to at most $\sim -12\,\mathrm{K}$. When in-28 cluding the results of previous studies, we find that relating SO₂ to any other param-29 eter is inconsistent across models compared to the relationships between AOD, RF, and 30 temperature anomaly ERF, and GMST. Thus, we expect the largest uncertainty in model 31 codes to relate to Finally, we find that the peak RF approaches a limiting value, and that 32 the peak temperature response follows linearly, effectively bounding the temperature anomaly 33 to at most $\sim -12 \,\mathrm{K}^{reviewer1:}$ [L25] the chemistry and physics of aerosol evolution. 34

Plain Language Summary

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Volcanic eruptions can have a significant impact on the Earth's climate. Eruptions large enough that reviewer1: [L31] the aerosols that form from the gases they emit reach the stratosphere cause a cooling effect by reflecting sunlight. Typically, an eruption is measured by its impact on the opacity of the stratosphere and the change in the reviewer1: [L33] energy balance at the top of the atmosphere when the surface temperature is held fixed. The two measures are often assumed to be linearly related, but the linearity is tested only against eruptions useful only for eruptions of sizes seen in the last two millennia. We use a coupled climate model to simulate the impact of eruptions of sizes up to the

largest known eruptions. The smallest eruptions we simulate are still large enough to cause 44 global climate effects. We find a clear In addition to a non-linear relationship for cruptions 45 larger than the ones seen in the past two millennia. Our between energy imbalance and 46 stratospheric opacity with increasing eruption magnitude, our simulations and support-47 ing data shows that the eruption latitude significantly influences the development of the 48 relationship between energy imbalance and stratospheric opacity with time after the erup-49 tion. Additionally, we find evidence that the peak energy imbalance reaches a limit, and 50 that the peak temperature response follows linearly with the peak energy imbalance, also 51 reaching a limiting value. 52

1 Introduction

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Effective radiative forcing (RFERF) and stratospheric aerosol optical depth (AOD) 54 are crucial metrics representing the energy imbalance at top-of-the-atmosphere (TOA) 55 and when ocean and sea-ice is held fixed and the stratospheric opacity due to aerosol scat-56 tering, respectively. They While calculation and estimation of forcing has not always had 57 an agreed-upon methodology, including both instantaneous radiative forcing (IRF) describing 58 the radiative forcing at TOA, similar to ERF but where ERF also accounts for rapid adjustments, 59 and stratospherically adjusted radiative forcing (RF) (Forster et al., 2016). Still, ERF 60 and AOD are extensively used to quantify the impact of major volcanic eruptions. The 61 , and a general assumption of a linear dependency of RF-ERF on AOD is commonly adopted 62 (Myhre et al., 2013; Andersson et al., 2015), and applying. Applying such a linear re-63 lationship has yielded reasonably accurate estimates in climate model simulations of volcanic eruptions (Mills et al., 2017; Hansen, Nazarenko, et al., 2005; Gregory et al., 2016; Marshall et al., 2020; Pitari et al., 2016). Yet, a wide spread in the estimated aerosol forc-66 ing efficiencies (RF-ERF normalised by AOD) exists among studies, spanning approx-67 imately from $\sim -15\,\mathrm{Wm}^{-2}\mathrm{AOD}^{-1}$ (Pitari et al., 2016) to (Myhre et al., 2013) reviewer1: [L55] 68 $\sim -25 \, \mathrm{Wm}^{-2} \mathrm{AOD}^{-1}$ (Hansen, Sato, et al., 2005). Additionally, these estimates are pre-69 dominantly based on small eruptions with AOD values up to at most ~ 0.7 . 70 Although H₂O, N₂, and CO₂ are the most abundant gases emitted by volcanoes 71

Although H₂O, N₂, and CO₂ are the most abundant gases emitted by volcanoes (Robock, 2000), sulphur species such as SO₂ provide a greater influence due to the comparatively high background concentrations of the former gases in the atmosphere. The transformation of SO₂ molecules through reactions with OH and H₂O leads to the formation of sulphuric acid (H₂SO₄) (Robock, 2000)(Pinto et al., 1989; Zhao et al., 1995)

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, which scatters sunlight thereby elevating planetary albedo and reducing the RFERF.
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       As the conversion from SO<sub>2</sub> to H<sub>2</sub>SO<sub>4</sub> reviewer1: [L63] occurs over weeks reviewer1: [L63] (Pinto et al., 1989; Zhao et al.,
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       , the peak RF H_2SO_4 burden experiences a slight delay from the eruption's peak SO_2
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      injection. The lifetime of the H_2SO_4 aerosols in the stratosphere depends on various fac-
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       tors, including aerosol size (Rampino & Self, 1982; Pinto et al., 1989; Marshall et al., 2019)
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       , latitude (Marshall et al., 2019; Toohey et al., 2019), volcanic plume height (Marshall
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       et al., 2019), aerosol size (Marshall et al., 2019), the quasi-biennial oscillation phase (Pitari
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       et al., 2016) and the season of the year (determining to which hemisphere aerosols are
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       transported) (Toohey et al., 2011, 2019). In the case of tropical eruptions, aerosols are
       typically transported poleward in the stratosphere and descend back to mid-latitude tro-
       posphere within one to two years (Robock, 2000). Upon descending below the tropopause,
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       these aerosols are readily removed by wet deposition (Liu et al., 2012).
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            reviewer1: [L75] Before the current era of significant anthropogenic climate forcing, volcanic
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       eruptions were the primary forcing mechanism dictating Earth's climate variability during
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       the Holocene period (Schurer et al., 2013). Despite this substantial impact, few climate-
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      model experiments have included volcanic forcing when simulating climate evolution dur-
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      ing the Holocene (Sigl et al., 2022), likely implying an exaggerated positive forcing (Gregory
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       et al., 2016; Solomon et al., 2011). This absence of persistent cooling is one of several
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      factors that have been suggested to contribute to the common disparity between sim-
       ulated and observed global warming (Andersson et al., 2015). Despite extensive atten-
       tion on understanding the way volcanic eruptions influence climate, questions regard-
      ing aerosol particle processes—such as growth and creation rates when OH is scarce—
       remain unanswered (e.g. Robock, 2000; Zanchettin et al., 2019; Marshall et al., 2020, 2022)
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       (e.g., Zanchettin et al., 2019; Marshall et al., 2020, 2022; McGraw et al., 2024). These
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       processes impact aerosol scattering efficiency and potentially the RF-ERF to AOD re-
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      lationship. Marshall et al. (2020) observe higher aerosol forcing efficiency in post-eruption
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      years 2 and 3 compared to year 1, and attribute this post-eruption increase in aerosol
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      forcing efficiency to strong spatial concentration in the initial year and subsequent dis-
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       tribution of aerosols over a larger area. This spatial redistribution increases the albedo
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      per global mean AOD thereby causing a stronger RF ERF to AOD ratio (Marshall et
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      al., 2020).
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            Previous studies of both Mt. Pinatubo (Mills et al., 2017; Hansen, Nazarenko, et
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       al., 2005) and volcanoes within the instrumental era (Gregory et al., 2016) have been used
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to estimate the relationship between the RF-ERF energy imbalance and change in AOD
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        caused by volcanic eruptions. While Myhre et al. (2013) employ a formula scaling RF
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        ERF by AOD to obtain -25 \,\mathrm{Wm}^{-2}\mathrm{AOD}^{-1}, recent literature reports estimates down to
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        reviewer1: [L94]-19.0\pm0.5\,\mathrm{Wm^{-2}AOD^{-1}} reviewer1: (\mathrm{Gregory\ et\ al., 2016})\ \mathrm{and}\ -18.3\pm1.0\,\mathrm{Wm^{-2}AOD^{-1}}
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        (Mills et al., 2017) [These are stratospheric aerosol simulations, not volcano simulations.]. Syn-
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        thetic volcano simulations in Marshall et al. (2020) yield a scaling factor of -20.5\pm0.2\,\mathrm{Wm}^{-2}\mathrm{AOD}^{-1}
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        across an ensemble of 82 simulations featuring varying injection heights and latitudes
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        of volcanic emissions, with injected SO_2 ranging from 10 to 100 \, \mathrm{Tg}(SO_2).
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A similar simulation setup, albeit with notable differences, was conducted by Niemeier and Timmreck (2015), involving an ensemble of 14 levels of injected sulphur spanning between $1 \text{ Tg(S)yr}^{-1} (2 \text{ Tg(SO}_2)\text{yr}^{-1})$ and $100 \text{ Tg(S)yr}^{-1} (200 \text{ Tg(SO}_2)\text{yr}^{-1})$. These geoengineering simulations maintained continuous sulphur injections, running until a steady sulphur level was achieved. Results indicated an inverse exponential relationship between RF and injected SO₂ maximum forcing and reviewer1: [L104] annually injected SO₂, converging to $-65\,\mathrm{Wm}^{-2}$ (Eq. 1). Even the $100\times\mathrm{Mt}$. Pinatubo super-volcano supereruption simulation by Jones et al. (2005), which obtained a peak RF-ERF of $-60 \,\mathrm{Wm}^{-2}$, is below the suggested limit of -65 Wm⁻². reviewer1: [L107] Moreover, Timmreck et al. (2010) find a peak ERF anomaly of $-18 \,\mathrm{Wm}^{-2}$ from a $1700 \,\mathrm{Tg}(\mathrm{SO}_2)$ eruption simulation, which corresponds well with the function estimated by Niemeier and Timmreck (2015) at an annual injecting rate of 1700 Tg(SO₂)yr⁻¹. Several studies have demonstrated a linear relationship of approximately $-20 \,\mathrm{Wm}^{-2}\mathrm{AOD}^{-1}$ between RF ERF and AOD, although substantial variability exists in the slope among studies (Mills et al., 2017; Hansen, Nazarenko, et al., 2005; Gregory et al., 2016; Marshall et al., 2020; Pitari et al., 2016). Moreover, a time-after-eruption dependence on the RF ERF to AOD ratio is found in Marshall et al. (2020), whereas Niemeier and Timmreck (2015) revealed a non-linear relationship between RF ERF and injected SO₂ rate. Thus, a consensus on the relationship between injected SO₂, AOD, and RF-ERF has yet to be established.

One avenue that has garnered considerable attention is comparing the magnitude of volcanic or volcano-like forcings to increased CO₂ levels. Several studies explore the connection between volcanic forcing and the climate sensitivity to a doubling of CO₂ (Boer et al., 2007; Marvel et al., 2016; Merlis et al., 2014; Ollila, 2016; Richardson et al., 2019; Salvi et al., 2022; Wigley et al., 2005). The comparison of forcing from volcanoes and CO₂ aims to mitigate the large uncertainty in estimates of the sensitivity of the real cli-

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mate system. Inferring climate sensitivity from volcanic eruption events has been attempted
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       as a way to constrain the sensitivity (Boer et al., 2007) by assuming that volcanic and
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       CO<sub>2</sub> forcings produce similar feedbacks (Pauling et al., 2023). Earlier studies suggest
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       the potential for constraining equilibrium cilmate sensitivity (ECS) using volcanoes (Bender
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       et al., 2010), provided that ECS is constrained by effective radiative forcing (ERF) rather
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       than instantaneous radiative forcing (IRF), as ERF accounts for rapid atmospheric ad-
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       justments in contrast to IRF (Richardson et al., 2019) (C. J. Smith et al., 2018; Richardson et al., 2019; Marshall
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       . However, other studies refute this approach, pointing out that different sensitivities of
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       volcanic forcing and CO<sub>2</sub> doubling seem to exist (Douglass et al., 2006), or that constrain-
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       ing the ECS by ERF lacks accuracy due to the precision of climate simulations (Boer
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       et al., 2007; Salvi et al., 2022). Although ERF offers a more suitable indicator of forc-
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       ing than IRF (Marvel et al., 2016; Richardson et al., 2019), more recent studies conclude
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       that ECS cannot be constrained from volcanic eruption events (Pauling et al., 2023).
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            reviewer1: [L135] Employing eruptions in the large to supercruption size enhances the
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       signal-to-noise ratio without necessitating an extensive and computationally expensive
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       ensemble, and as such, is a tempting shortcut to try and mimic a large ensemble of smaller
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       volcanic eruptions. However, the AOD, RF, and temperature ERF, and GMST signa-
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       tures are not necessarily a simple scaling of that of smaller volcanic eruptions. Previous
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       studies have simulated super-volcanoes supercruptions using AOD as the input forcing,
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       where the AOD was that of Mt. Pinatubo scaled by a factor of one hundred (Jones et
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       al., 2005). This approach may More recent studies find this approach will yield incor-
       rect results, both because the peak of the AOD may be too small or too is likely to be
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       too big, but also because the evolution of the AOD could be inappropriate due to the
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       aerosol size (Timmreck et al., 2009, 2010). Likewise, a different AOD evolution may be
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       found from similar size eruptions, but at different latitudes (Schneider et al., 2009; Marshall et al., 2020; Zhuo et a
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       . To investigate this issue, our simulations are based on four levels of injected SO<sub>2</sub> cov-
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       ering three orders of magnitude and the inclusion of one high latitude eruption of the
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       second largest injected SO<sub>2</sub> case.
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We conducted ensemble simulations of volcanic eruptions in the Community Earth System Model Version 2 (CESM2) coupled with the Whole Atmosphere Community Climate Model Version 6 (WACCM6). The ensembles span four different levels of injected SO₂: 26 Tg(SO₂), 400 Tg(SO₂), 1629 Tg(SO₂) and 3000 Tg(SO₂). Details regarding the experimental setup are provided in section 2. Our findings reveal non-linear RF-ERF

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to AOD dependencies for medium_large to super-volcano size eruptions. Additionally, we observe a time-dependent variation in the RF-ERF to AOD ratio, detailed in section 3 and discussed in section 4. Furthermore, our data, along with insights from previous studies, suggest that the RF-ERF dependency on injected SO_2 identified by Niemeier and Timmreck (2015) acts as a lower boundary. Our conclusions are presented in section 5.

2 Method

2.1 Model

We use the CESM2 (Danabasoglu et al., 2020) in conjunction with the WACCM6 (Gettelman et al., 2019) and the fully dynamical ocean component Parallel Ocean Program version 2 (POP2) (R. Smith et al., 2010; Danabasoglu et al., 2020). The atmosphere model was run at a nominal 2° resolution with 70 vertical levels in the middle atmosphere (MA) configuration.

The WACCM6 version employed in the MA configuration uses the three mode version of the Modal Aerosol Module (MAM3) (Gettelman et al., 2019), a simplified and computationally efficient default setting within the Community Atmosphere Model version 5 (CAM5) (Liu et al., 2016), as described in Liu et al. (2012). The MAM3 was developed from MAM7 and features the modes Aitken, accumulation, and coarse (Liu et al., 2016), and further updated to simulate stratospheric sulfate aerosol from volcanic and non-volcanic emissions (Mills et al., 2016).

2.2 Simulations

reviewer1: [L157]

We are using the coupled model version BWma1850 component setup to run the CESM2 with a fully dynamic ocean component to get estimates of the GMST, and an accompanying fixed sea-surface temperature version, fSST1850, providing estimates of the ERF and AOD. The applied fSST1850 is not from a standardised component setup of CESM2 but is instead explicitly specified as 1850_CAM60%WCCM_CLM50%BGC-CROP_CICE% PRES_DOCN%DOM_MOSART_CISM2%NOEVOLVE_SWAV_TEST. The component setup BWma1850 and fSST1850 differ in that the latter uses a prescribed sea-ice (CICE -> CICE%PRES), a prescribed data ocean (POP2%ECO%DEP -> DOCN%DOM) and a stub wave component instead of the full Wave Watch version 3 (WW3 -> SWAV).

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The important input data used in the model simulations are injected SO<sub>2</sub> in units
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       of teragrams (Tg(SO_2)), used to simulate volcanic eruptions. ERF is calculated as the
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       combined (short wave and long wave) all-sky TOA energy imbalance, where the CESM2
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       provide the output variables "net solar flux at the top of the model" (FSNT) and "net
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       longwave flux at the top of the model" (FLNT). Thus, ERF_* = FSNT - FLNT, and taking
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       the difference between volcanic forcing simulations and a control simulation gives the final
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       estimate of ERF (ERF = ERF<sub>VOLG</sub> - ERF<sub>CONTROL</sub>) (Marshall et al., 2020). The ERF
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       calculation uses the fSST1850 component setup, which is also used to obtain all other
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       simulation output fields except from GMST which uses BWma1850. The AQD is obtained
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       from the output variable "stratospheric aerosol optical depth 550 nm day night" (AODVISstdn),
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       while GMST is saved by CESM2 to the variable "reference height temperature" (TREFHT).
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       The analysis of this work is performed using these four variables.
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             Appendix A provides a description of the simulation setupand utilised output variables.
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       Table 1 summarises the simulations, encompassing four SO<sub>2</sub> injection magnitudes and
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       up to four seasons: 15 February, 15 May, 15 August, and 15 November. reviewer2: [MC2]
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       The magnitudes vary over three orders of magnitude, or as introduced in Schmidt and Black (2022)
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       across volcano-climate index values 3 to 6: 26 \operatorname{Tg}(SO_2), 400 \operatorname{Tg}(SO_2), 1629 \operatorname{Tg}(SO_2), and
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       3000 \operatorname{Tg}(SO_2).
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             The smallest eruption case, C2W $\$S26$, is similar in magnitude as compared to events
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       like Mt. Pinatubo (\sim 10-20 \,\mathrm{Tg}(\mathrm{SO}_2); Timmreck et al., 2018) and Mt. Tambora (\sim 56.2 \,\mathrm{Tg}(\mathrm{SO}_2);
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       Zanchettin et al., 2016). (~144–170 Tg(SO<sub>2</sub>); Vidal et al., 2016) reviewer1: [L178] The intermediate
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       case, S400, resembles the magnitude of the Samalas eruption in 1257 (\sim 144-170 \,\mathrm{Tg}(\mathrm{SO}_2); Vidal et al., 2016)
       however injecting about twice of the estimated SO<sub>2</sub>, while the second largest and largest
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       eruption cases, C2W<sup>↑</sup> and C2W<sup>↑↑</sup>S1629 and S3000, is in the likely range of the Young
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       Toba Tuff (YTT) eruption supercruption occurring about 72 000 yr ago (100–10 000 Tg(SO<sub>2</sub>);
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       Jones et al., 2005). All eruptions were situated at the equator (0 °N, 1 °E) with SO<sub>2</sub> reviewer1: [L182]
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       injected from 18 km to 20 km altitude with a linear ramp; 25% between 17.5 km and 18.5 km,
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       50\% between 18.5 \,\mathrm{km} and 19.5 \,\mathrm{km}, and the last 25\% between 19.5 \,\mathrm{km} and 20.5 \,\mathrm{km}. Col-
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       lectively, the four tropical eruption cases C2W\, C2W\, and C2W\\, sad C2W\\\, S26, S400,
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       $1629, and $3000 are referred to as C2WTrop STrop. An additional high-latitude erup-
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       tion ensemble, labelled \frac{\text{C2WN}}{\text{S}_{1}} \frac{\text{S}_{2}}{\text{S}_{2}}, of the same injected SO<sub>2</sub> magnitude as \frac{\text{C2W}}{\text{C}_{2}}
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       S1629 was simulated at 56 °N, 287.7 °E with a six-month separation (15 February and
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       15 August) between the two simulations.
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Table 1. $^{reviewer1:}$ [Table 1] Simulations done with the CESM2 a

Ensemble name	$Tg(SO_2)$	Lat [°N]	Lon [°E]	Alt [km]
C2W↑↑ <u>S3000</u>	3000	0	1	18-20
<u>C2WN</u> ↑ <u>\$1629N</u>	1629	56	287.7	18 – 20
$\frac{\text{C2W}}{\text{S1629}}$	1629	0	1	18-20
$ \underbrace{\text{C2W}}_{\text{S400}} $	400	0	1	18 - 20
C2W ↓ <u>S26</u>	26	0	1	18 - 20

^aThe ensembles C2WN↑ and C2W↑ have the same cruption magnitude, but while C2W↑ is located at the equator, C2WN↑ is located at a high northern latitude. C2W↑↑, C2W— and C2W↓ are located at the equator, but with different magnitudes compared to C2W↑. The three smallest tropical ensembles have four members, indicated by the number of cruption months, while the northern latitude and the extra large super-volcano ensemble consists of two members.

^aThe ensembles S1629 is located at the equal S26 are located at the three smallest tropical months, while ensembles

3 Results

3.1 Analysis of the time series

Figure 1 presents time series of global mean AOD, RF, and surface air temperature ERF, and GMST. The black lines represent the medians across the ensembles, while shading indicates the 5th to 95th percentiles. The four distinct forcing magnitudes (C2W\$\dagger\$, C2W\$\dagger\$, and C2W\$\dagger\$, S400, S1629, and S3000) outlined in table 1 have been used. The time series in Fig. 1 are normalised by setting the peak value to unity, defined based on the peak of a fit from a Savitzky-Golay filter of 3rd order and a one-year window length (Savitzky & Golay, 1964).

is not found in the AOD time series. C2W\S26 peaks at an earlier time, but also spends more time around the peak and as such decays at a later time post-eruption. Likewise, C2W—S400 has a faster rise and slower decay compared to C2W\S1629, but where both peak at a similar time. C2W\ and C2W\ have S1629 and S3000 have reviewer1: [L204] similar normalised AOD developments, but where C2W\ S3000 show a slightly faster decay from the peak.

The timescale of the perturbation of AOD and RF-ERF is shorter than that of the temperature GMST. While the AOD and RF-ERF time series return to their equilibrium state within roughly three years, the temperature GMST time series remain heavily perturbed three years post-eruption. Even when running the simulations for 20 years post-eruption, the temperature GMST time series are still decaying.

3.2 RF ERF dependency on AOD

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We next focus on the development of the AOD and RF-ERF time series relative to each other. Similar comparisons were conducted in Gregory et al. (2016, their Fig. 4) and Marshall et al. (2020, their Fig. 1), with RF-ERF plotted against AOD. Figure 2 displays annual mean values from the five simulation cases in table 1; the small eruption case (C2W+S26) as blue downward-pointing triangles, the intermediate eruption case (C2W—S400) as orange thick diamonds, the large tropical eruption case (C2W↑S1629) as green upward-pointing triangles, the extra large eruption case (C2W\S3000) as small pink upward-pointing carets, and the large northern hemisphere eruption case (C2WN+\$\frac{\color{1}}{2}S1629N) as brown upward-pointing three-branched twigs. Also shown are the data from Gregory et al. (2016, Fig. 4, black crosses from HadCM3 sstPiHistVol) as grey crosses labelled G16 (described in Appendix B, section B3). Additionally, the estimated peak values from the Mt. Pinatubo and Mt. Tambora eruptions are plotted as a black star and plus, while the peak from the Jones et al. (2005) simulation is shown as a pink square labelled J05. Finally, red circles represent the peak values obtained from the C2W eruption cases. The straight lines are the same as shown by Gregory et al. (2016). The full data range is shown in Fig. 2a while Fig. 2b highlights a narrow range, focusing on the C2W \$26 case.

The annual mean data from the Pinatubo-like C2W\$_\$26 case in Fig. 2b have RF

ERF values as a function of AOD that follow almost the same constant slope as the G16

data. However, in Fig. 2a we observe that the stronger eruptions lead to dissimilar re-

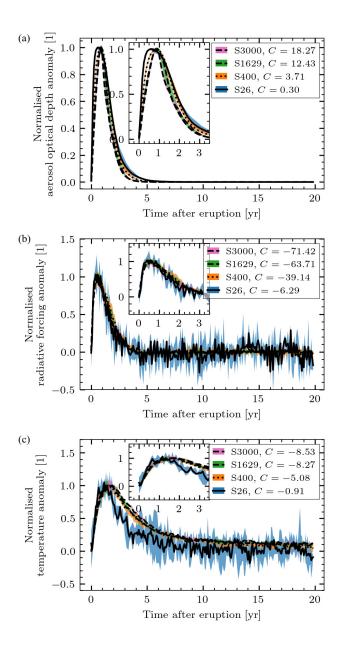
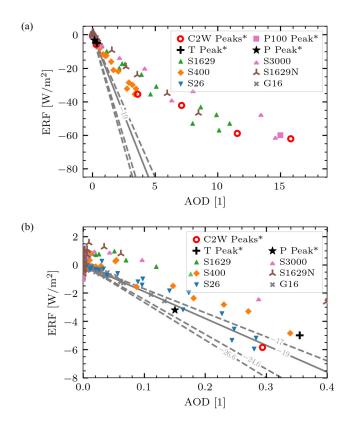


Figure 1. AOD (a), RF-ERF (b) and temperature GMST response (c) time series to the four tropical volcanic eruption cases, $C2W \downarrow S26$, C2W - S400, $C2W \uparrow S1629$, and $C2W \uparrow S3000$. The time series have been normalised to have peak values at unity, where C is the normalisation constant. Black lines indicate the median across the ensembles, while shading marks the 5th and 95th percentiles.



sponses in AOD and RF, where C2W—ERF, where S400 seems to follow close to a -10 slope and C2W \uparrow -S1629 is closer to a -5 slope. The peak values (red circles) suggest a non-linear dependence, while within each eruption strength (same colour) the annual mean values fall relatively close to a straight line.

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To investigate the time dependence of the ratio between RF ERF and AOD, we present seasonal means of this ratio in Fig. 3. The plot shows the eruption cases given in table 1, as well as the tropical eruptions from Marshall and Smith (2020) (6 of 82 erup-

tions), labelled M20 and described in Appendix B, section B2. The C2W↑-\$1629 case is similar to C2W↑↑-\$3000 as indicated in table 2, but is not shown in the plot to better highlight C2WN↑better highlight \$1629N. In Fig. 3a, lines are linear regression fits to the seasonal means across all ensemble members, summarised in table 2. Shaded regions are the standard deviation around the seasonal means. A similar shading is plotted in Fig. 3b, but where the regression fits have been omitted for clarity. As the AOD and RFERF time series start from zero, the ratio from the first season is not included. Likewise, after three years both time series are almost fully equilibrated (Fig. 1a,b). The data is further divided into two periods; a pre-peak period where the peak of both the AOD and the RFERF is included (consisting of the first post-eruption year), and a post-peak period for the decaying part (consisting of the second and third post-eruption years).

Although the ratio changes across the eruption magnitudes, we find that all the tropical cases follow a positive slope during the pre-peak period, as seen in Fig. 3a and described in table 2. The northern latitude case in C2WN+S1629N shows a much flatter slope compared to C2WTrop STrop and M20. The distinction between the slopes from the tropical and non-tropical cases is perhaps more clear in Fig. 3b and corresponding rows in table 2. Again, C2WN+S1629N shows an almost flat slope compared to the tropical cases. During the post-peak period, more noise is introduced, but a weak tendency of negative slopes is found among the tropical cases, as well as in the C2WN+S1629N case up to the last season where the noise is also the largest.

Marshall et al. (2020, their Fig. 1c,d) present results that demonstrate a time-dependent relationship in the conversion between AOD and RFERF. They obtain an RF-ERF to AOD ratio with a negative slope when comparing the first post-eruption year to the second and third. As such, Marshall et al. (2020) find that, on average, the aerosol forcing efficiency increases during the first two to three post-eruption years. This phenomenon is explained by Marshall et al. (2020) as the aerosols initially being spatially confined to the hemisphere where the eruption occurred. Subsequently, during the second and third years, they spread globally, resulting in a higher global-mean albedo per AOD and consequently a stronger RF-ERF per AOD ratio with time. However, as noted above, a decrease in aerosol forcing efficiency is found when analysing the M20 data with seasonal resolution during the pre-peak period (first year post-eruption) while constraining the ensemble to only include eruptions within -10 to 10 °N. reviewer1: [L269] The post-peak period shows an increasing aerosol forcing efficiency, and during the full first three post-eruption

Table 2. Slope and standard deviation for the data in Fig. 3^a

Figure	Ensemble name	Pre-peak	Post-peak
	C2WN↑- <u>\$1629N</u>	0.45 ± 1.15	1.51 ± 1.45
	C2W↑↑_S3000	3.38 ± 0.97	-2.74 ± 0.77
3a	C2W↑ <u>\$1629</u>	3.85 ± 0.52	-3.29 ± 0.60
	C2W — <u>\$400</u>	4.36 ± 0.82	-3.37 ± 0.59
	C2W↓ <u>S26</u>	3.64 ± 2.41	-1.41 ± 3.25
	M20	6.34 ± 1.77	-0.36 ± 1.33
3b	C2WN↑-S1629N	0.08 ± 0.20	0.27 ± 0.26
	C2W↑↑ S3000	0.86 ± 0.25	-0.70 ± 0.19
	C2W↑ <u>S1629</u>	0.75 ± 0.10	-0.64 ± 0.12
	C2W — <u>\$400</u>	0.43 ± 0.08	-0.34 ± 0.06
	C2W↓ <u>S26</u>	0.18 ± 0.12	-0.07 ± 0.16
	M20	0.33 ± 0.07	-0.02 ± 0.08

^aThe regression fits in the top half of the table are for Fig. 3a, while the bottom half is for Fig. 3b. The columns "pre-peak" and "post-peak" refer to the two periods as shown in Fig. 3. The ensembles are the same as those given in table 1, in addition to the 6 tropical eruptions from the 82 member ensemble in Marshall et al. (2020).

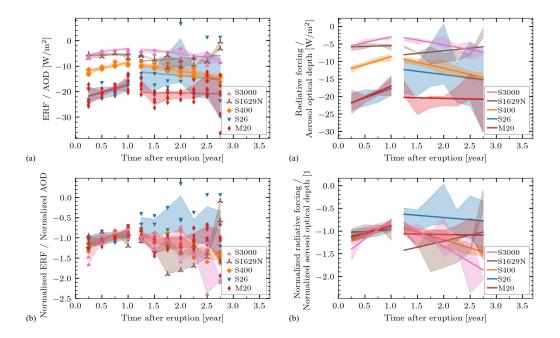


Figure 3. reviewer1: [Figure 3](a): The ratio of RF-ERF to AOD, with time-after-eruption on the horizontal axis. Straight lines indicate linear regression fits and are described in table 2, while shaded regions are the standard deviation across the ensembles for each season. Regression fits and shadings are made for the pre-peak and post-peak periods. (b): Same as in (a), but where the underlying AOD and RF-ERF time series have been scaled to have peak values at unity. Shown are data from table 1 along with tropical eruptions from M20.

years (pre-peak and post-peak), both the tropical subset and the full M20 data yield an increasing efficiency, as expected. Likewise, the first three post-eruption years of the C2W, and C2WN↑ S400, S3000, and S1629N cases show a weak negative slope and thus an increasing efficiency, while C2W S26 shows an elevated post-peak ratio as seen in Fig. 3b.

We also note that while the aerosol forcing efficiency is decreasing for tropical M20 data in the pre-peak period, the full dataset shows increasing efficiency. This is in line with what we find from C2WN↑S1629N, which is the only eruption case that does not show a clear aerosol forcing efficiency decrease during the pre-peak period.

3.3 Parameter scan

In Fig. 4, we compare the peak values of all investigated CESM2 output param-eters against each other as well as to injected SO_2 . For our, grouped into tropical cases (C2WTrop), STrop) and the high-latitude case (S1629N). We also include data from Marshall et al. (2020) (M20); Jones et al. (2005) (J05); Timmreck et al. (2010) (T10); English et al. (2013) (E13); Niemeier and Timmreck (2015) (N15); Otto-Bliesner et al. (2016) (OB16); Brenna et al. (2020) (B20); Osipov et al. (2020) (Os20); and McGraw et al. (2024) (McG24). A description of the climate models used is presented in table C1. Additionally, peak values from Mt. Pinatubo (P) and Mt. Tambora (T) are shown for reference.

For STrop, we observe in Fig. 4a an almost linear yet notably weakening relationship between AOD peak values and injected SO₂. The latitude also plays a role in the magnitude of the AOD perturbation, evident from C2WN†S1629N. This weak yet notable latitude dependence aligns with findings by Marshall et al. (2019), indicating that 72% of the AOD variance can be attributed to injected SO₂, while latitude accounts for only 16% of the variance. Peak values from their data (82 simulations) plotted as red thin diamonds display a similar pattern, with AOD exhibiting close to linear dependence on injected SO₂, but with latitude introducing a spread in AOD. Peak values from Mt. Pinatubo (P) and Mt. Tambora (T) are shown for reference, along with align well with simulation data, while peak values from Jones et al. (2005) labelled other simulations of large SO₂ magnitudes (J05and Timmreck et al. (2010) labelled. T10. The J05 is a simulation of a super-volcano based on a 100 times scaling of the AOD from Mt. Pinatubo, while T10 is a simulation of the YTT cruption based on SO₂ injections. E13, B20, Os20) show, a larger spread, specifically to weaker AOD response.

In Fig. 4b, RF-ERF plotted against injected SO₂ (with the absolute value of RF-ERF on the y-axis) indicates a substantial damping effect on RF-ERF as injected SO₂ increases for the C2W-STrop data, in agreement with results from Otto-Bliesner et al. (2016), labelled OB16. The OB16 data come from a 2500 year long simulation using historic volcanoes as the only external forcing. The analysis details of OB16 can be found in Appendix B, section B1. Despite the model complexity difference, Otto-Bliesner et al. (2016)'s simulations using Community Earth System Model version 1 (CESM1) with a low-top atmosphere (CAM5) produce RFs-ERFs comparable to our findings. Both B20 and McG24 align well with STrop, and while B20 uses the same climate model, McG24

uses the GISS ModelE2.1 but where a fixed aerosol effective radius of $R_{\rm EFF} = 0.6\,\mu{\rm m}$ was used. This $R_{\rm EFF}$ is at the lower end of their simulations, which is shown by McGraw et al. (2024) to produce the most extreme ERF and GMST perturbations.

Niemeier and Timmreck (2015) conducted simulations of continuous sulphur injections up to $200 \,\mathrm{Tg}(\mathrm{SO}_2)\mathrm{yr}^{-1}$ in the ECHAM5's middle atmosphere version (Giorgetta et al., 2006) with aerosol microphysics from HAM (Stier et al., 2005). They observed an RF-ERF dependence on SO_2 injection rate following an inverse exponential, which converges to $-65 \,\mathrm{Wm}^{-2}$, depicted in Fig. 4b as the stippled pink line labelled N15 and given as;

$$\Delta R_{\text{TOA}} = -65 \,\text{Wm}^{-2} \frac{e^{-\left(\frac{2246 \,\text{Tg(S)yr}^{-1}}{x}\right)^{0.23}}}{\exp \left[-\left(\frac{2246 \,\text{Tg(S)yr}^{-1}}{x}\right)^{0.23}\right]} . \tag{1}$$

Both our simulations and OB16 exhibit a notably faster increase than this exponential relationship. The results by N15, on which Eq. 1 is based, are all averages over at least three years of steady sulphur burdens, substantially longer than the time it takes for RF ERF to reach peak values after an eruption. Combined with their lack of a full chemistry model (Niemeier & Timmreck, 2015), a direct comparison between Eq. 1 to peak RF-ERF values (occurring about one year post-eruption) may not reflect the same chemical and physical processes. In Eq. 1, x represents S, while the axis shows values of SO₂, thus halving of the SO₂ values on the axis gives the appropriate shape of Eq. 1 as a function of S.

With these caveats in mind, we observe that even though most simulations exhibit a notably faster increase than the exponential relationship, T10's results closely align with the function described in Eq. 1. Starting with an initial input of 850 Tg(S) (equivalent to 1700 Tg(SO₂), representing the YTT eruption), their estimated AOD led to a peak RF-ERF of -18 Wm⁻², depicted as a pink filled circle in Fig. 4b. The results from T10 came from a simulation using the MPI-ESM climate model, driven by AOD data from the HAM aerosol model. This alignment likely stems from the utilization of the same aerosol microphysical model in both Timmreck et al. (2010) and Niemeier and Timmreck (2015), as well as the application of similar climate models, MPI-ESM and ECHAM5, respectively. The relationship between climate model families and their implications are further described in Appendix C. Notably, the peak values from M20 fit well within an upper boundary defined by C2WTrop-STrop and OB16, and a lower boundary defined

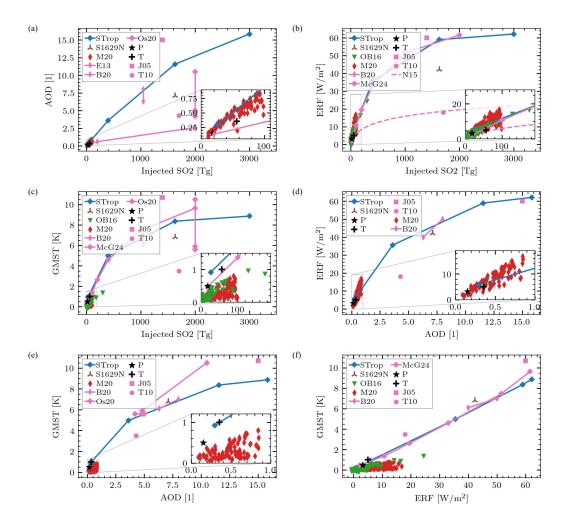


Figure 4. reviewer1: [Fig. 4] Peak values of (a) AOD, (b) RFERF, and (c) temperature anomaly GMST as a function of injected SO₂. (d) RF ERF and (e) temperature anomaly GMST as a function of AOD. (f) Temperature anomaly GMST as a function of RFERF. Blue diamonds labelled C2WTrop STrop represent tropical cases (C2W+S26, C2W—S400, C2W+S1629, C2W+S3000), the brown three-branched twig signifies the C2WN+S1629N case, and green downward triangles denote OB16 data from Otto-Bliesner et al. (2016). The red thin diamonds labelled M20 display the Marshall and Smith (2020) data. Black star and plus indicate Mt. Pinatubo and Mt. Tambora estimates based on observations. The pink square labelled J05 refers to the one-hundred times Mt. Pinatubo super-volcano from Jones et al. (2005), and the pink disk labelled T10 represents the YTT super-volcano from Timmreck et al. (2010). The pink dashed line labelled N15 is from Niemeier and Timmreck (2015), indicating the function in Eq. 1.

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by Eq. 1. Eruptions closer to the equator within M20 align with data points near the
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       upper boundary, whereas eruptions at more extreme latitudes tend to yield weaker peak
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       RF-ERF values, closer to the lower boundary. Importantly, none of the eruption sim-
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       ulations shown in Fig. 4b exceeded the upper threshold of -65\,\mathrm{Wm}^{-2} as suggested in
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       Eq. 1.
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            Figure 4c illustrates the response of temperature GMST against injected SO<sub>2</sub>. The
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       increase in temperature GMST response with injected SO<sub>2</sub> decreases for higher injected
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       SO<sub>2</sub>, showing a similar relationship between C2WTrop, C2WN↑, and STrop, S1629N,
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       OB16, B20, and McG24. Notably, T10 and J05 exhibit respectively much weaker and
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much stronger temperature GMST responses to injected SO_2 than C2WTrop. STrop.

while Os20 cover a wide range in GMST for the same injected SO_2 of $2000\,\mathrm{Tg}(SO_2)$. In

Os20 they removed a single mechanism at the time, with four experiments with GMST

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between $5.5\,\mathrm{K}$ and $6\,\mathrm{K}$, and one at $\sim 10.5\,\mathrm{K}$. For this outlier, feedback on photochemistry

due to aerosols had been switched off (Osipov et al., 2020). T10 has a maximum temperature

GMST anomaly of only $-3.5 \,\mathrm{K}$ for their $1700 \,\mathrm{Tg}(\mathrm{SO}_2)$ eruption, while J05 records a sub-

stantially larger maximum temperature GMST anomaly of -10.7 K. Since the M20 ex-

periment was conducted with prescribed sea-surface temperatures (Marshall et al., 2020),

preventing the temperature GMST from being fully perturbed, reviewer1: [L338] we do not

focus on the M20 data in the GMST plots but include them for completeness.

In Fig. 4d, we revisit the relationship between RF-ERF and AOD, focusing on peak values rather than annual and seasonal averages. As previously discussed, the RF-ERF to AOD ratio displays weaker slopes than previous studies, with the C2W-STrop peak values not conforming to a linear trend. The relationship between RF-ERF and AOD suggests potential substantial dependencies on the model and its input parameters, such as latitude, but most notably to an inherent non-linear RF-ERF dependence on AOD. Both the G16 data in Fig. 2 and the J05 data originate from the same climate model. Similarly to what we find from the C2W-STrop data, the ratio is much stronger for small eruptions in the industrial era (G16) compared to the super-volcano eruption (J05).

In Fig. 4e, we again find that the response of the C2WTrop-STrop data decreases with injected SO₂increasing AOD, this time in temperature GMST anomaly. Additionally, both the C2WN↑ and the J05 cases S1629N and B20 align well with C2WTrop, with

the STrop, with T10ease, and J05 and Os20 following a similar dependence, albeit somewhat weaker and stronger, respectively.

Finally, in Fig. 4f, we compare the temperature and RF GMST and ERF responses.

Both C2WTrop STrop and OB16 show a near-linear relationship between temperature and RF. The C2WTrop GMST and ERF. The STrop data indicate a steeper slope, implying stronger temperature GMST perturbations as compared to OB16. However, potential biases exist in the values from the analysis of OB16, as outlined in Appendix B, section B1. This, along with considerable noise, results in the analysis of OB16 temperature GMST anomalies being less reliable. As in Fig. 4c, the C2WN↑ case along with both the T10 and All other cases (S1629N, J05cases, T10, B20, McG24) closely follow the temperature to RF dependence of C2WTropGMST to ERF dependence of STrop.

The almost linear relationship between AOD and injected SO₂ for the C2WTrop

STrop data in Fig. 4a suggests a comparable trend for RF-ERF versus injected SO₂ in

Fig. 4b, as seen for RF-ERF versus AOD in Fig. 4d. For the same reason, we expect Fig. 4e to show a similar pattern for C2WTrop STrop as observed in Fig. 4c.

This relationship, along with the functional relationships between all other parameters shown in Fig. 4, are illustrated in Fig. 5. There, $^{reviewer1:}$ we show that from assuming a linear dependency of AOD on injected SO₂ [why is this being assumed when it has been shown this isn't very valid?] $^{reviewer1:}(ax+b)$, and of GMST on ERF (cx+d) [It seems as if x is referring to different quantities in the different equations. Much more attention to detail needed if this mathematical framework is to be explained in sufficient detail.], we must have that f, g, h, and k all have the same functional form, where $f: SO_2 \to RFf: SO_2 \to ERF$, $g: AOD \to T$, $h: SO_2 \to T$, and $k: AOD \to RFk: AOD \to ERF$. From this, we deduce that f(x) = k(ax+b) and h(x) = f(cx+d) = g(ax+b), and finally that h(x) = k(acx+ad+b), concluding that f, g, h, and k have the same functional form.

3.4 Climate sensitivity estimate

As previously mentioned, the J05 experiment is similar to $C2W\uparrow$ concerning RF S1629 concerning ERF values, yet differ in both AOD and temperature GMST. At the same time J05 is similar to $C2W\uparrow\uparrow$ S3000 in AOD and RFERF. To investigate this discrepancy, we here conduct a comparison between the J05 climate feedback parameter α (where $s = 1/\alpha$ is the climate sensitivity parameter) with our climate resistance, de-

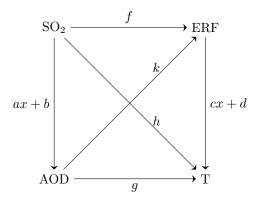


Figure 5. Diagram describing the functional relationships of the parameters shown in Fig. 4.

noted as ρ , and the transient climate response parameter (TCRP) $1/\rho$ (where TCS = $F_{2\times CO_2} \times TCRP$ is the transient climate sensitivity and $F_{2\times CO_2}$ is the forcing due to a doubling of pre-industrial CO₂ concentration). , a duration too short reviewer! [L378] As the forcing of volcanic eruptions last for too short time for $F = \rho T$ to remain valid, an alternative approach using a time-integral form introduced by Merlis et al. (2014) is applied:

$$\int_0^\tau F dt = \rho \int_0^\tau T dt \tag{2}$$

$$\rho = \frac{\int_0^\tau F dt}{\int_0^\tau T dt}.$$
 (3)

If the upper bound of the integral, τ , is sufficiently large, so that the upper ocean heat content is the same at t=0 and $t=\tau$ (Merlis et al. (2014) used $\tau=15\,\mathrm{yr}$), this approach agrees with $F=\rho T$ for long-term forcing (Gregory et al., 2016). Additionally, we note that the climate resistance and the climate feedback parameter are associated with the ocean heat uptake efficiency (κ) through $\rho=\alpha+\kappa$ (Gregory et al., 2016).

The climate feedback parameter estimated by Jones et al. (2005) is $\alpha \simeq 4 \, \mathrm{Wm}^{-2} \mathrm{K}^{-1}$, exceeding twice the value obtained by Gregory et al. (2016) in their simulations of Mt. Pinatubo using the same HadCM3 climate model. We determine the climate resistance using the integral-form computation outlined in Eq. 3 and adopting $\tau = 20 \, \mathrm{yr}$. The estimated climate resistance from the three tropical simulation cases (with four in each en-

Table 3. Estimated climate resistance and $TCRP^a$

Simulation type	$\rho[\mathrm{Wm^{-2}K^{-1}}]$	1/ ho
C2W↑ <u>S1629</u>	2.21 ± 0.05	0.45 ± 0.01
C2W— $S400$	2.51 ± 0.06	0.40 ± 0.01
C2W↓_S26_	2.9 ± 0.6	0.36 ± 0.07
Total	2.5 ± 0.4	0.41 ± 0.05

^aEstimates are based on ensembles with four members and $\tau = 20 \,\mathrm{yr}$ using Eq. 3.

semble) converges to $\rho = 2.5 \pm 0.4 \, \mathrm{Wm}^{-2} \mathrm{K}^{-1}$, and TCRP values of $1/\rho = 0.41 \pm 0.05 \, \mathrm{KW}^{-1} \mathrm{m}^2$, as reported in table 3, and is therefore assumed to be a good estimate of α .

Importantly, our estimate agrees well with G16, while the J05 estimate of $\alpha \simeq 4\,\mathrm{Wm}^{-2}\mathrm{K}^{-1}$ is still notably higher. Since the temperature GMST perturbation obtained by J05 was larger than in any of our CESM2 cases, it indicates that the forcing used by J05 must be stronger. reviewer1: [L398] The peak value of the J05 ERF is similar to the S1629 case, and as such, the overall stronger forcing must originate from the development of the forcing time series rather than the peak value. This is in line with recent studies investigating the effect of different aerosol effective radius. McGraw et al. (2024) find that from supereruptions, it is possible to achieve even a warming of the GMST. However, as our model results align well with the results of McGraw et al. (2024) using a small aerosol effective radius of $R_{\rm eff} = 0.6\,\mu\rm m$, we expect the peak GMST from CESM2(WACCM6) to be close to a lower bound, and as such that the even colder GMST perturbation of Jones et al. (2005) is too extreme.

4 Discussion

Figures 2, 3, and 4d demonstrate that as the AOD exceeds approximately 1.0, the linear RF-ERF dependence of approximately $-20 \,\mathrm{Wm^{-2}AOD^{-1}}$ no longer holds. The sublinear increase in RF-ERF with injected SO₂ in Fig. 4b for large eruptions is consistent with previous results from simulations using similar climate models of smaller historic eruptions (G16) and of super-volcanoes supereruptions (J05). Such a change in ratio has been attributed to larger eruptions, injecting more SO₂, leading to larger aerosols, and hence less effective radiation scattering, thereby reducing the RF-ERF for the same

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injected SO<sub>2</sub> (English et al., 2013; Timmreck et al., 2010, 2018). Similarly, previous studies
have suggested a two-thirds power law relationship between peek AOD and injected SO<sub>2</sub>
for eruptions larger in magnitude than than the Mt. Tambora eruption (Crowley & Unterman, 2013; Metzner et a
. Furthermore, CESM(WACCM) has been shown to simulate smaller aerosols than most
other climate models (Clyne et al., 2021), resulting in an increased AOD peak value and
e-folding time (Zanchettin et al., 2016; Clyne et al., 2021). Thus, the sublinear relationship
for AOD and ERF to injected SO<sub>2</sub> from CESM2(WACCM6) is likely an upper bound.
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The non-linear relationship between peak RF-ERF and AOD values is a strong signature in both Figs. 2 and 3. Across eruptions of the same strength, the ratio stays relatively constant, leading to a close to $-10\,\mathrm{Wm^{-2}AOD^{-1}}$ slope for C2W—S400 and a $-5\,\mathrm{Wm^{-2}AOD^{-1}}$ slope for C2W↑ and C2W↑ \$1629 and \$3000. Still, a non-linear development in the RF-ERF to AOD ratio is found across all tropical eruptions. Similar to the results of Marshall et al. (2020), we find in C2W , C2W↑, and C2W↑↑ \$400, \$1629, and \$3000 that the post-peak period (second and third post-eruption years) has a stronger aerosol forcing efficiency compared to the pre-peak period (first post-eruption year). The post-peak period of C2W↓ \$26 is elevated as compared to the pre-peak period, resulting in a decreasing aerosol forcing efficiency from the first to the second and third post-eruption years, in contrast to the other tropical eruptions.

Focusing on the pre-peak period, we find tropical eruptions to differ from eruptions at high latitudes. During the pre-peak period, all tropical eruptions show a decreasing aerosol forcing efficiency, while no significant change in the RF-ERF to AOD ratio is found from the C2WN+S1629N case. The full M20 dataset indicates an increasing aerosol forcing efficiency also during the pre-peak period, contrasting the decreasing efficiency found from their tropical eruptions and supporting the latitudinal dependence we find with C2WN+S1629N. While we find a linear relationship to be a useful approximation of RF-ERF dependence on AOD for eruptions similar to or smaller than Mt. Pinatubo, additional factors must be considered for larger eruptions. These factors, such as OH scarcity and aerosol growth, influence reflectance and their gravitational pull, substantially impacting both AOD and RF-ERF evolution, is highlighted by Timmreck et al. (2010). The large difference in ratio found when comparing eruption magnitudes suggests that injected SO₂ is crucial when estimating the time-average of the RF-ERF to AOD ratio. However, latitude and, in par-

ticular, aerosol dispersion are more influential in determining the post-eruption evolution of the ratio, particularly during the pre-peak period.

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We find that the suggested upper threshold from Eq. 1 is not violated by any eruption simulation, and most notably that the temperature GMST peak value follow the RF-ERF trend in reaching a limiting value. The C2WTrop STrop cases follow a close to linear temperature dependency on RF, with the GMST dependency on ERF, with S1629N, J05, T10, and C2WN \uparrow -B20, and McG24 all aligning close to the same slope. The linear relationship between temperature and RF-GMST and ERF is the strongest dependence found between the parameters in Fig. 4, and a strong signature across both eruption magnitudes and latitudes, but also across highly different climate models. Thus, from a maximum RF-ERF of $-65 \, \mathrm{Wm}^{-2}$, we expect temperature GMST anomalies to reach at most $\sim -12 \, \mathrm{K}$, in support of English et al. (2013) who suggested that large eruptions can be self-limiting.

The biggest spread in the data shown in Fig. 4 is found when relating injected SO₂ to any of the three output parameters. As the amount of injected SO₂ increases, both AOD, RF, and temperature ERF, and GMST across models have a big spread. The AOD to injected SO₂ relationship is consistent within similar models, even when comparing simulations of volcanic eruptions (Timmreck et al., 2010) and continuous injection of SO₂ (Niemeier & Timmreck, 2015), but has a wide spread at high values of injected SO₂ across model families (Figs. 4a,b,c). Comparatively, the RF-ERF (Fig. 4d) and temperature GMST (Fig. 4e) as a function of AOD, as well as temperature GMST as a function of RF-ERF (Fig. 4f), demonstrate a smaller spread across models. Marshall et al. (2019, 2020, 2021) use a code with seven log-normal modes to simulate aerosol mass and number concentrations, along with an atmosphere-only configuration of the UM-UKCA with prescribed sea-surface temperatures and sea-ice extent (Marshall et al., 2019). This approach is in contrast with CESM2, operating as an Earth System Model, but with a simpler aerosol chemistry model in the MAM3. The family of models to which M20 is based is different from that of C2W and OB16, and also different from the T10 and N15, as described in Appendix C. Based on Fig. 4, we find the model family to be pivotal in determining the estimated AOD and RF-ERF magnitudes from injected SO₂, whereas the various models generally demonstrate more consistency in representing RF-ERF from AOD.

Timmreck et al. (2010) highlights that for sufficiently large eruptions, OH radicals are too scarce, which limits SO₂ oxidation. The AOD peak in the YTT simulation of T10 occurs six months after Mt. Pinatubo's peak. This aligns with our results, as illustrated in Fig. 1a, where C2W+S26 shows an earlier AOD peak compared to C2W -, C2W+, and C2W+S400, S1629, and S3000. While the peak RF-ERF value of T10 occurs 7-8 months post-eruption, similar to C2WSTrop, the J05 peak anomaly occurs one year post-eruption. Additionally, as Jones et al. (2005) obtains a climate feedback parameter larger than both what Gregory et al. (2016) found for the same climate model and larger than the climate resistance obtained here from C2WSTrop, we conclude that such a simple approach of scaling the AOD of smaller cruptions to represent larger cruptions is insufficient. Moreover, having a small ensemble of large cruptions to represent smaller cruptions is also insufficient when simulating from injected SO₂, as both AOD and temperature evolution are found to develop differently, their estimated GMST is likely too extreme.

5 Summary and conclusions

We consider five medium to super-volcano sized eruption ensembles ensembles of Mt. Pinatubo-sized to supercruption sized events and compare them to previously reported results. We find the commonly adopted RF-ERF dependence on AOD of $\sim -20\,\mathrm{Wm}^{-2}\mathrm{AOD}^{-1}$ to be representative for Mt. Pinatubo-sized eruptions. Larger eruptions, with one to two orders of magnitude larger injections of SO₂, are found to have an RF-ERF dependence on AOD closer to $\sim -10\,\mathrm{Wm}^{-2}\mathrm{AOD}^{-1}$ and $\sim -5\,\mathrm{Wm}^{-2}\mathrm{AOD}^{-1}$. A shallower slope for larger eruptions is also consistent with peak values from previous studies of super-volcanoes supercruptions.

The time-after-eruption dependence of the ratio between RF-ERF and AOD is found to weaken with time, resulting in a decreasing aerosol forcing efficiency in the pre-peak period. The effect is found across all eruption sizes, but only the tropical cases show a clear trend. The high-latitude case displays an almost constant efficiency with time. These results agree with a reanalysis of the tropical data in Marshall and Smith (2020). Thus, these findings provide strong supporting evidence that latitude is generally significant in determining the aerosol forcing efficiency, particularly as a function of time-after-eruption. These findings emphasise the complexity of volcanic impacts on climate, demonstrating significant differences in climatic response depending on eruption magnitude and latitude.

We find that the AOD peak arrives later for larger eruptions than for smaller ones, and also that larger eruptions produce a sharper peak in the AOD time series. The RF ERF time series are similar across all eruption sizes, and while the smallest eruption experiences a faster temperature GMST decay, the larger eruptions produce time series indistinguishable in development for both RF and temperature ERF and GMST. Thus, a simple scaling of the AOD or temperature GMST time series from a smaller eruption is insufficient in representing that of larger volcanic eruptions.

Considering injected SO₂ and the peak values of AOD and RFERF, a large spread is found across model families in Fig. 4. Improving the consistency between model families in how the chemistry and physics of SO₂ and H₂SO₄ are represented is an important step in enhancing the accuracy of simulated volcanic eruptions' influence on climate by models. More simulations of larger volcanic eruptions with injected SO₂ greater than 200 Tg(SO₂) would provide useful information for a more precise determination of the RF-ERF to AOD ratio in the non-linear regime. This would also serve as a useful test to check if a comparison between SO₂ injection events and continuous SO₂ injection is reasonable. Introducing a spread in latitude similar to the Marshall and Smith (2020) dataset would allow for better comparison between eruptions across all latitudes and the suggested lower limit following Eq. 1, describing a situation of aerosol saturation.

Appendix A Simulation set up and output set up

Input files used in the simulations were created by modifying the file available at http://svn.code.sf.net/p/codescripts/code/trunk/ncl/emission/createVolcEruptV3.ncl, using a Python package available on GitHub at https://github.com/engeir/volcano-cooking or through the Python Package Index (PyPI). The package is available both as a library and a Command Line Interface (CLI), and is used to create volcanic eruptions with a specified amount of SO₂ that is injected over six hours at a given latitude, longitude, and altitude. All volcanic SO₂ files are created from a shell script by setting the eruption details in a JSON file that is read by the volcano-cooking CLI at a fixed version, ensuring a reproducible experiment setup.

We are using the coupled model version BWma1850 component setup to run the CESM2, and an accompanying fixed sea-surface temperature version, fSST1850, to obtain estimates of the RF. The applied fSST1850 is not from a standardised component setup but is instead

explicitly specified as . The component setup BWma1850 and fSST1850 differ in that the latter uses a prescribed sea-ice (CICE -> CICE%PRES), a prescribed data ocean (POP2%ECO%DEP -> DOCN%DOM) and a stub wave component instead of the full Wave Watch version 3 (WW3 -> SWAV).

The important input data used in the model simulations are injected SO_2 in units of teragrams ($Tg(SO_2)$), used to simulate volcanic cruptions. RF is calculated as the combined (short wave and long wave) all-sky TOA energy imbalance, where the CESM2 provide the output variables "net solar flux at the top of the model" (FSNT) and "net longwave flux at the top of the model" (FLNT). Thus, $RF_* = FSNT - FLNT$, and taking the difference between volcanic forcing simulations and a control simulation gives the final estimate of RF ($RF = RF_{VOLC} - RF_{CONTROL}$) (Marshall et al., 2020). The RF calculation is based on £SST1850, hence this outline specifically describes how to calculate ERF as opposed to IRF, which instead is the difference between the ERF and the sum of all rapid atmospheric adjustments (Marshall et al., 2020; C. J. Smith et al., 2018). The AOD is obtained from the output variable "stratospheric acrosol optical depth 550 nm day nigth" (AODVISstdn), while global temperature is saved by CESM2 to the variable "reference height temperature" (TREFHT). The analysis of this work is performed using these four variables.

During analysis, one outlier was found in the ensemble representing C2W\$\\$26\$, specifically in the temperature GMST time series. This ensemble member was the February 15, 1850, eruption, which was changed in favor favour of a February 15, 1851, eruption in the C2W\$\\$, C2W\$\\$, and C2W\$\\$\\$\$S26, S400, and S1629 ensembles. For completeness, the February 15, 1850, eruption is still included in the online archive.

Appendix B External data

B1 Otto-Bliesner data analysis

Data from Otto-Bliesner et al. (2016) are the original input data of injected SO₂ as used in their model simulations, along with RF and temperature ERF and GMST output data. The injected SO₂ can be found at https://www.cesm.ucar.edu/working-groups/paleo/simulations/ccsm4-lm. Only the peak values of the SO₂ dataset were used in the analysis. Output variables are available at www2.cesm.ucar.edu/models/experiments/LME.

Since the OB16 dataset contains a five-member ensemble, the final RF and temperature ERF and GMST time series used were ensemble means. A single control simulation time series is used to remove seasonal dependence from the temperature GMST, where the control simulation is averaged into a climatology mean. Further, a drift in the temperature GMST is removed by subtracting a linear regression fit. RF-ERF has seasonality removed in the Fourier domain.

The time of an eruption is found based on a best attempt at aligning the SO₂ time series with both the RF-ERF time series and the temperature GMST time series. The RF and temperature ERF and GMST peak values are taken as the value of the time series at the time of an eruption according to the SO₂ time series. Missing the true peak means the found peaks will be biased towards lower values. However, instances where eruptions occur close in time will contribute a bias to higher values. These biases contribute to a greater uncertainty related to OB16 in Figs. 4b,c,f.

B2 Marshall data analysis

Data used to compute the M20 values were from Marshall and Smith (2020), available at https://doi.org/10.5285/232164e8b1444978a41f2acf8bbbfe91. As each file includes a single eruption, peak values of AOD, RF, and temperature ERF, and GMST were found by applying a Savitzky-Golay filter of third order and one-year window length, and choosing the maximum value (Savitzky & Golay, 1964).

B3 Gregory data analysis

Data used to compute G16 values were kindly provided by Jonathan Gregory (personal communication). The full 160-year-long time series were further analysed by computing annual means.

Appendix C Model families

The model used here was the CESM2 with the WACCM6 atmosphere in the MA configuration. The MA configuration uses the MAM3 (Gettelman et al., 2019), a simplified and computationally efficient default setting within the CAM5 (Liu et al., 2016), as described in Liu et al. (2012). The MAM3 was developed from MAM7, consisting of the seven modes Aitken, accumulation, primary carbon, fine dust, fine sea salt, coarse

Table C1. Model code family relations^a

Family relation	Model name	
CDCM1 - CDCM1 CAME - CDCM0	CESM1	
$CESM1 \rightarrow CESM1\text{-}CAM5 \rightarrow CESM2$	CESM2	<u>B20</u> ,
${\rm HadCM3} \rightarrow {\rm HadGEM1} \rightarrow$	HadCM3	
$\operatorname{HadGEM2} \to \operatorname{HadGEM3} \to \operatorname{UM-UKCA}$	UM-UKCA	
$\text{ECHAM5} \rightarrow \text{ECHAM6} \rightarrow \text{MPI-ESM}$	ECHAM5	
ECHAM3 → ECHAM0 → MF1-ESM	MPI-ESM	
GISS-E2.1	GISS-E2.1	

^aOverview of various model codes grouped into families according to the model code genealogy map by Kuma et al. (2023), with each table entry also indicating the specific model code used in the referenced papers of this study.

^aOverview of various genealogy map by Ku model code used in the WACCM3, only simul OB16, B20 and this compared to the code used in the code

dust, and coarse sea salt. Instantaneous internal mixing of primary carbonaceous aerosols with secondary aerosols and instantaneous ageing of primary carbonaceous particles are assumed by emitting primary carbon in the accumulation mode (Liu et al., 2016). As dust absorbs water efficiently and is expected to be removed by wet deposition similarly to sea salt, fine dust is merged with fine sea salt into the accumulation mode and coarse dust is merged with coarse sea salt into a coarse mode. The coarse mode will quickly revert to its background state below the tropopause (Liu et al., 2012). Consequently, MAM3 features the three modes Aitken, accumulation, and coarse (Liu et al., 2016).

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The CESM2 is an ancestor of CESM1 used by OB16. They belong to a different model family than both the HadCM3 (J05 and G16) and the UM-UKCA (M20), which is an extended version of HadGEM3 (Dhomse et al., 2014), and an ancestor of HadCM3. A third model family is represented through ECHAM5 (N15) and MPI-ESM (T10), where the latter is related to the former via the ECHAM6. A summary of the model code genealogy is in table C1, based on the model code genealogy map created by Kuma et al. (2023).

8 Acronyms

- AODVISstdn "stratospheric aerosol optical depth 550 nm day night"
- AOD stratospheric aerosol optical depth
- 701 CAM5 Community Atmosphere Model Version 5
- 702 **CESM1** Community Earth System Model Version 1
- 703 **CESM2** Community Earth System Model Version 2
- 704 **ECS** equilibrium climate sensitivity
- 705 **ERF** effective radiative forcing
- FLNT "net longwave flux at the top of the model"
- FSNT "net solar flux at the top of the model"
- 708 IRF instantaneous radiative forcing
- MAM3 three mode version of the Modal Aerosol Module
- $_{710}$ MA middle atmosphere
- POP2 Parallel Ocean Program Version 2
- T12 ERF effective radiative forcing
- TCRP transient climate response parameter
- TOA top-of-the-atmosphere
- 715 **TREFHT** "reference height temperature"
- 716 WACCM6 Whole Atmosphere Community Climate Model Version 6
- 717 YTT Young Toba Tuff

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Open Research Section

The direct output data of CESM2 are too large to be easily archived and transferred. Instead, data generated directly from output fields of CESM2 are made available in a NIRD Research Data Archive (?, ?)(Enger, 2024b), and were generated using scripts available at https://github.com/engeir/cesm-data-aggregator. Analysis scripts are available at GitHub (https://github.com/engeir/code-to-radiative-forcing-by-super-volcano-eruptions) and is published to Zenodo (?, ?)(Enger, 2024a). Source code used to generate CESM2 input files are available at https://github.com/engeir/cesm2-volcano-setup.

Acknowledgments

- The simulations were performed on resources provided by Sigma2 the National Infrastructure for High Performance Computing and Data Storage in Norway.
- This work was supported by the Tromsø Research Foundation under Grant Number 19_SG_AT.
- Thanks to both Maria Rugenstein and Martin Rypdal for valuable discussions. We would also like to thank the authors of Gregory et al. (2016), Otto-Bliesner et al. (2016), and Marshall and Smith (2020) for making their data available.

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