Initial <u>atmospheric</u> conditions control transport of volcanic volatiles, forcing and impacts

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Abstract. Volcanic eruptions impact the climate and environment. The volcanic forcing is determined by eruption source parameters, including mass and composition of volcanic volatiles, eruption season, eruption latitude and injection altitude. Moreover, initial <u>atmospheric</u> conditions of the climate system play an important role in shaping the volcanic <u>forcing and</u> response. However, our understanding of the combination of these factors, the distinctions between tropical and extratropical

- 5 volcanic eruptions and the co-injection of sulfur and halogens remains limited. Here, we perform ensemble simulations of volcanic eruptions at 15° N and 64° N in January, injecting 17 Mt of SO₂ together with HCl and HBr at 24 km altitude, considering different initial conditions of the El Niño-Southern Oscillation, Quasi-Biennial Oscillation, and polar vortex. Our findings reveal that initial atmospheric conditions control the transport of volcanic volatiles from the first month and modulate the subsequent latitudinal distribution of sulfate aerosols and halogens. This results in different volcanic forcing,
- 10 surface temperature and ozone responses over the globe and Northern Hemisphere Extratropics (NHET) among the model ensemble members with different initial <u>atmospheric</u> conditions. NH extratropical eruptions exhibit a larger NHET mean volcanic forcing, surface cooling and ozone depletion compared to tropical eruptions. However, tropical eruptions lead to more prolonged impacts compared to NH extratropical eruptions, both globally and in the NHET. The sensitivity of volcanic forcing to varying eruption source parameters and model dependency is discussed, emphasizing the need for future multi–model studies
- 15 to consider the influence of initial conditions and eruption source parameters on volcanic forcing and subsequent impacts.

1 Introduction

Explosive volcanic eruptions can inject sulfur dioxide (SO₂), halogens, ash, and water vapor into the stratosphere, causing significant perturbation of the Earth system. Stratospheric sulfate aerosols formed from injected SO₂ reflect incoming solar radiation and absorb longwave radiation, cooling the surface and warming the stratosphere (Robock, 2000; Timmreck, 2012).

A lot of studies have been conducted on the explosive eruption of Mt. Pinatubo in 1991 (Bluth et al., 1992; Guo et al., 2004; Robock, 2000; Ukhov et al., 2023). However, large uncertainties still exist in quantifying the amount of SO₂ injected into the stratosphere based on different observational (Bluth et al., 1992; Grant et al., 1992; Guo et al., 2004) and modelling (Dhomse et al., 2014; Jones et al., 2016; Mills et al., 2016; Niemeier et al., 2009; Quaglia et al., 2023; Stenchikov et al., 2021) studies. To reach the best agreement with Pinatubo volcanic forcing observations, models have used a wide range of SO₂ injections from 10

- to 20 Tg (Timmreck et al., 2018). Atmosphere–aerosol models are mostly used, either with prescribed sea surface temperature (SST), such as MAECHAM5–HAM (Niemeier et al., 2009) and UM–UKCA (Dhomse et al., 2014), or in a fully coupled configuration with interactive atmosphere, ocean, sea–ice, chemistry and with a high model top, such as CESM–WACCM (Mills et al., 2016). Even when injecting the same SO₂ mass, models simulate different sulfate burden (Marshall et al., 2018) and stratospheric aerosol optical depth (SAOD) (Clyne et al., 2021; Zanchettin et al., 2016). Clyne et al. (2021) and Mills et al.
- 30 (2017) argued that interactive OH chemistry is essential to accurately simulate the evolution and lifetime of volcanic aerosols. Quaglia et al. (2023) showed that with interactive aerosol microphysics, different stratospheric transport in different models is the main reason for their disagreement with Pinatubo observations.

Previous modelling studies on volcanic impacts mostly considered sulfur as the only volcanic volatile injection, ignoring the potential of volcanic halogens reaching the stratosphere (Krüger et al., 2015; Kutterolf et al., 2013, 2015; von Glasow et al.,

- 35 2009; WMO, 2018). The few studies that simulated volcanic eruptions with co-injection of sulfur and halogens showed different volcanic forcing, ozone and climate responses compared to eruptions with sulfur-only injection (Brenna et al., 2019, 2020; Klobas et al., 2017; Lurton et al., 2018; Ming et al., 2020; Staunton-Sykes et al., 2021). However, these studies modelled different strengths of volcanic eruptions with different atmospheric background conditions. A more systematic model study investigating impacts of sulfur and halogen-rich tropical and extratropical eruptions is still lacking.
- 40 Different source parameters, including mass and composition of volcanic volatiles, eruption season, eruption latitude and eruption altitude, contribute to the source of large uncertainties that shape the forcing and impact of volcanic eruptions (Marshall et al., 2019; Metzner et al., 2014; Toohey et al., 2011, 2019; Zhuo et al., 2014, 2021)(Kravitz and Robock, 2011; Marshall et al., A key question lies in understanding how eruption latitude affects the forcing and impact of volcanic eruptions. Tropical eruptions were considered to have larger climate impact than extratropical eruptions (IPCC, 2013; Schneider et al., 2009). However,
- 45 ice cores and tree ring reconstructions indicate that compared to historical tropical eruptions, extratropical eruptions led to larger climate impacts per unit volcanic stratospheric sulfate injection over the Northern Hemisphere (NH), which was supported by analysis of MAECHAM5–HAM model simulations (Toohey et al., 2019). Nevertheless, the results of this study are limited with regard to the aerosol and climate response as the ocean temperature and sea ice were prescribed, next to as well as the missing interaction of atmospheric chemistry and QBOQuasi–Biennial Oscillation (QBO). Using the fully coupled model
- 50 MPI–ESM, Zhuo et al. (2021) simulated stronger NHET surface cooling after NH extratropical volcanic eruptions compared to tropical eruptions. However, this study used prescribed SAOD as volcanic forcing, which does not explicitly simulate the chemical, microphysical, and dynamical processes of the aerosols that play an important role in shaping the volcanic forcing and subsequent climate impact.

Post-eruption responses are affected by pre-eruption meteorological and climate initial conditions, including the initial

55 states of the The evolution of stratospheric volcanic materials and associated radiative forcing has been suggested to depend on the state of the atmosphere at the time of the eruption. The initial spread of volcanic clouds depends on the wind direction at the time of eruption, as evidenced by the different volcanic cloud distribution observed a few weeks after 1982 El Niño–Southern Oscillation (ENSO), Quasi–Biennial Oscillation (QBO) and polar vortex. The pre–eruption ENSO state modulates the post–eruptionENSO responses to volcanic eruptions and the surface winter warming pattern (Coupe and Robock, 2021; Khodri et al., 2017; Pausata et al., 2016.

- 60 Chichón and 1991 Pinatubo eruptions (Robock, 2000). In the stratosphere, the QBO dominates the tropical circulation with alternating easterly and westerly winds , which plays an important role in the distribution of stratospheric acrosols and chemical constituents (Baldwin et al., 2001). Unlike ENSO and QBO, studies mostly considered the response of the polar vortex to tropical volcanic eruptions (Charlton-Perez et al., 2013; Toohey et al., 2014), but rarely considered how the initial state of the polar vortex affects the volcanic forcing by influencing the distribution and evolution of acrosols from volcanic
- 65 eruptions (Baldwin et al., 2001). In particular, differences in the impact of initial conditions on volcanic forcing and subsequent impacts between tropical and extratropical eruptions are unknownStratospheric aerosols experience greater lofting and tropical confinement under easterly shear conditions in the QBO region, as opposed to when a westerly shear is present (Trepte and Hitchman, 1992) . In the extratropical stratosphere, the initial stability of polar vortex (PV) largely modulates evolution and distribution of volcanic materials (Fuglestvedt et al., 2024), which also relates to the seasonal cycle of the Brewer Dobson Circulation (BDC,
- 70 Butchart, 2014; Toohey et al., 2011) and seasonal variation of OH concentration at different latitudes (Fuglestvedt et al., 2024; Toohey et al., 2019). Most of these variabilities operate on scales ranging from hourly to interannually, collectively shaping the evolution and distribution of volcanic materials and subsequently determining volcanic forcing.

The volcanic forcing plays an important role in determining the post-eruption climate response. The initial climate state such as the pre-eruption El Niño-Southern Oscillation (ENSO) state modulates the post-eruption ENSO responses to volcanic

75 eruptions and the surface winter warming pattern (Coupe and Robock, 2021; Khodri et al., 2017; Pausata et al., 2016, 2020). For addressing the role of the initial climate state, the use of large model ensembles and multi model comparison is suggested (Zanchettin et al, 2022).

Here, we use a fully coupled Earth system model with interactive atmosphere, ocean, sea ice, and prognostic aerosol microphysics and atmospheric chemistry to simulate Pinatubo–magnitude eruptions at 15° N and 64° N in January in a pre–industrial

- atmosphere. We are particularly interested in how eruptions of this Pinatubo-strength are affected by a QBO westerly phase, given the QBO disruption and prolonged easterly regime response to a tropical super eruption as modelled by Brenna et al. (2021). In addition, we apply different initial conditions of atmospheric conditions, under westerly QBO conditions, which include different ENSO and polar vortex states for our tropical and NH extratropical eruptions with sulfur and halogen injections. We also conduct sensitivity tests with sulfur injections only and in eruption season of July. In a related study by
- 85 Fuglestvedt et al. (2023)Fuglestvedt et al. (2024), we analyze the effects of initial polar vortex conditions on the aerosol evolution, volcanic forcing and deposition of NH extratropical eruption at 64° N. Here in In this study, we aim to understand address the following questions: What impacts do initial conditions have on How do initial atmospheric conditions influence transport of volcanic volatiles and volcanic forcing after tropical eruptions^{co}-injecting, particularly in light of the co-injection of sulfur and halogens into the stratosphere? What differences do tropical and NH extratropical volcanic eruptions have on volcanic
- 90 forcing and subsequent climate and ozone impacts?

In the following, we describe the data and methods in Sect. 2, including the model description and experimental design. In Sect. 3, we present the results. Sect. 3.1 shows the transport of SO_2 and aerosol controlled by initial <u>atmospheric</u> conditions, which determines the volcanic forcing after tropical eruptions co–injecting sulfur and halogens. In Sect. 3.2, we compare the

aerosol evolution, volcanic forcing, climate and ozone impact of tropical and NH extratropical eruptions under different initial

95 atmospheric conditions. The results are discussed in Sect. 4. Finally, we give the summary and conclusions in Sect. 5.

2 Data and Methods

2.1 Model description

This study uses the Community Earth System Model, version 2 (CESM2; Danabasoglu et al., 2020) with the high-top version of the atmosphere the Whole Atmosphere Community Climate Model, version 6 (WACCM6; Gettelman et al., 2019). WACCM6
100 has a horizontal resolution of 0.95° latitude by 1.25° longitude, with 70 hybrid sigma-pressure levels extending from the Earth's surface to 6 × 10⁻⁶ hPa (~140 km altitude). The ocean component, Parallel Ocean Program version 2 (POP2; Smith et al., 2010) runs at a nominal 1° horizontal resolution, with 60 vertical layers to 5,500 meters in depth. The same grid is used by the Los Alamos National Laboratory sea ice model, version 5 (CICE5; Hunke et al., 2015). The land component is the Community Land Model Version 5 (CLM5; Lawrence et al., 2019), with the Community Ice Sheet Model Version 2.1 (CISM2.1; Lipscomb et al., 2019) as its land-ice component.

WACCM6 uses a comprehensive chemistry configuration for the troposphere, stratosphere, mesosphere and lower thermosphere (TSMLT). It includes 231 solution species and 583 chemical reactions with 150 photolytic, gas–phase and heterogeneous reactions relevant for the whole atmosphere (Gettelman et al., 2019). The chemical families include O_x , HO_x , NO_x , CIO_x , BrO_x and SO_x . Aerosols are treated using the modified Modal Aerosol Model version 4 (MAM4; Liu et al., 2016) which

110 is coupled to cloud microphysics (Liu et al., 2012) and incorporates prognostic stratospheric aerosols (Mills et al., 2016). Validation of SO₂ emission and radiative forcing of volcanic eruptions confirms the qualification of the model for simulating volcanic eruptions and stratospheric aerosol geoengineering (Mills et al., 2016, 2017). WACCM6 also includes an internally generated QBO (Gettelman et al., 2019).

For model intercomparison, we also adopt MAECHAM5–HAM (Stier et al., 2005) for sensitivity tests. MAECHAM5–HAM 115 is an aerosol–climate model with a T42 spectral truncation at $\sim 2.8^{\circ} \times 2.8^{\circ}$ spatial resolution and 39 vertical levels up to 0.01 hPa (~80 km). The model was run with free atmosphere and prescribed SSTs. Aerosol processes in the stratosphere are calculated by the aerosol microphysical module HAM (Niemeier et al., 2009; Toohey et al., 2011), which includes aerosol formation and growth via nucleation, condensation, accumulation and coagulation, sedimentation and final removal processes of wet and dry deposition. HAM uses a prescribed monthly mean OH field. This may affect the conversion of SO₂ to SO₄, which is dependent

120 on the local consumption of OH. In addition, we also use the modular volcanic forcing generator EVA (Easy Volcanic Aerosol, Toohey et al., 2016). It prescribes the SAOD and serves as the tool for compiling the volcanic forcing for experiments in the Paleoclimate Model Intercomparison Project 4 (Jungclaus et al., 2017; Sigl et al., 2015, 2022; Toohey and Sigl, 2017).

2.2 Experimental design

A spin–up run for 56 years is conducted under pre–industrial 1850s conditions, from which we pick six initial conditions

- 125 considering different El Niño–Southern Oscillation (ENSO), and polar vortex (PV) states with westerly Quasi–Biennial Oscillation (QBO) (atmospheric conditions to conduct ensemble runs (Table 1). For each condition, we select a range of ENSO with two cases in each of ENSO positive, neutral and negative states, and with varying QBO westerly and polar vortex strengths. The six cases otherwise represent a random sampling of meteorological variability. (see Section 2.3)to conduct ensemble runs (Table
- +). These baseline ensemble experiments include both volcanic sulfur and halogen injections. Tropical eruptions are located at
 15° N, 91° W (H–15N–Jan), simulating Central American Volcanic Arc (CAVA) eruptions, while NH extratropical eruptions
- are located at 64° N, 19° W (H–64N–Jan), simulating eruptions of the Katla volcanic system over Iceland. We use January as the base eruption season, as this is the default for historical volcanic eruptions when the eruption season is unknown in ice–core–based volcanic forcing reconstructions (Sigl et al., 2022; Toohey and Sigl, 2017). Following Toohey et al. (2019), we inject 17 Tg SO₂ at 24 km altitude, mimicking a Pinatubo–like eruption, but in a pre–industrial 1850s atmosphere taking the
- 135 co-injection of sulfur and halogens into account. The injected HCl and HBr masses are based on estimation of CAVA eruptions (Kutterolf et al., 2013, 2015), assuming a conservative injection efficiency of 10% for tropical eruptions to the stratosphere (Brenna et al., 2019; Krüger et al., 2015) but applied here for both eruption latitudes for comparability. Except for the baseline experiments, we also run simulations injecting only SO₂ at 15° N (S-15N-Jan) and 64° N (S-64N-Jan), to test the difference between volcanic eruptions with co-injection of sulfur and halogens and sulfur-only injection. We run a set of eruptions in
- 140 July at 15° N (H–15N–Jul and S–15N–Jul) and 64° N (H–64N–Jul and S–64N–Jul), to test the impact of eruption season on the volcanic forcing and related impacts. A control run lasts_lasting for 30 years without volcanic injection is conducted.

For comparison, we use model data from Toohey et al. (2019) simulating volcanic eruptions with 17 Tg SO₂ injection at 15° N in both January and July with MAECHAM5–HAM (ECHAM5–15N–Jan/Jul). For tropical eruptions, we calculate ensemble member-mean with five members each. For NH extratropical eruptions at 64° N, two additional runs from

145 ECHAM5–64N–Jan/Jul are available for comparison (this study). We also calculated the volcanic forcing from EVA (EVA–15N –Jan/Jul and EVA–64N–Jan/Jul).

The set of experiments is summarized in Table 1.

2.3 Analysis methods

We adopt the Oceanic Niño Index (ONI; https://www.climate.gov/news-features/understanding-climate/climate-variabilityoceanic-nino-index) to quantify the initial ENSO state. The ONI is calculated from a three-month running mean of the average sea surface temperature (SST) anomalies over the Niño 3.4 region covering 5° N-5° S and 170° W-120° W. The SST anomalies are calculated with respect to the 30-year mean of the control run without volcanic eruption. The QBO is calculated with the monthly equatorial mean zonal wind at 30 hPa between 2° N-2° S. We present both the ensemble members and the ensemble means of adopted variables to study the volcanic forcing and 155 subsequent impacts. Anomalies are calculated with respect to the 30-year mean of the control run. The ensemble standard deviation is calculated to indicate the spread of the baseline ensemble experiments.

3 Results

3.1 Initial atmospheric conditions control aerosol transport and volcanic forcing after tropical eruptions

Tropical eruptions have usually been thought to have a larger climate impact induce stronger surface cooling than extratropical
 eruptions, as SO₂ and formed aerosols are transported to both the Northern and Southern Hemispheres. Here, we examine how initial <u>atmospheric</u> conditions affect the transport and evolution of volcanic volatiles and subsequent volcanic forcing after tropical eruptions.

Figure 1a shows the first month latitudinal and vertical distribution of SO_2 concentration at 30 ppby after the 15° N tropical eruptions in January, which diverges among the baseline ensemble experiments. We have ordered the baseline experiments

- 165 . The contours reveal a range of dispersion of SO₂ across the six baseline ensemble members. In some simulations, SO₂ is strongly contained in the tropical pipe, with stronger vertical ascent, while in others, SO₂ is strongly mixed into the NH mid-latitudes. Accordingly, we labeled the ensemble members H1 to H6 according to less to more northward transport of volcanic volatiles, respectively based on the ratio of poleward versus upward transport, calculated by dividing the latitude- and altitude-weighted means of the first month SO₂ concentration over the regions poleward (20–30 hPa, 30–60° N) and upward
- 170 (5–15 hPa, 0–30° N) relative to the injection point (dash outlined in a). Note that this labeling differs from Fuglestvedt et al. (2023), who ordered Fuglestvedt et al. (2024), who labeled their ensemble members based on the stability of the polar vortex. During this boreal winter season, wave breaking is stronger in the winter hemisphere leading to strong mixing and distribution toward the NH high latitudes, which is evident especially in the first month, filaments of high SO₂ concentration are pulled towards NH mid–latitudes, particularly evident in H5 and H6 (Fig. S1). In contrast, when SO₂ is distributed more
- 175 equatorward, less SO₂ is distributed to the ensemble members with stronger upwelling exhibits stronger tropical confinement, and lesser distribution towards the NH, as shown exemplified especially in H1 - (Fig. 1a and S2).

The latitudinal distribution of SO₂ in the first month modulates the subsequent latitudinal distribution of SO₄. Figure 1b shows the latitudinal distribution of the sixth 10^{th} month SO₄ column burden for the six ensemble members. More H6 exhibits a greater transport of SO₄ is transported to the NH high latitudes between 30° N and 90° Nin H6, compared to the other members,

- 180 especially in particularly when contrasted with H1 and H2. Differences in the transport of SO₂ and SO₄ result in differences in cumulative volcanic forcing in the NH and SH high latitudes among the ensemble members (Fig. 1c). H6 shows a strong pronounced asymmetric volcanic forcing with large meridional forcing differences, i.e., e.g., 60 months cumulative SAOD up to 8.4 months between the NH and the SH, while. In contrast, H1 shows exhibits a relatively symmetric volcanic forcing between the hemispheres. Here, even with the same identical eruption source parameters, volcanic eruptions in the tropies result
- 185 in different symmetries of tropical volcanic eruptions give rise to differing degrees of hemispheric asymmetry of the volcanic forcing only due to different initial conditions. solely due to variations in initial atmospheric conditions. The inset scatter plot

clearly illustrates a correlation between the poleward/upward ratio of the first month SO2 and the hemispheric asymmetry of cumulative SAOD over 60 months. For tropical eruptions, the initial atmospheric conditions control the transport of SO₂ from the first month, to the formation and transport of SO₄ in the coming months and determine the latitudinal distribution of the sulfate aerosol and subsequent volcanic forcing.

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Figure 1d shows the latitudinal and vertical distribution of SO_2 and residual vertical velocity of the first post-eruption month for the six ensemble members. The SO_2 reaches higher altitudes but relatively less towards the NH extratropics in members H1 to H6. Notably, in H5 and H6, with greater transport of SO_2 to NH mid-latitudes, there is comparatively lower upward transport of SO_2 compared to the other members. Consequently, this leads to accelerated formation and transport of sulfate

- 195 aerosol towards the NH high latitudes in the following months (Fig. 1b). The residual vertical velocity (w*, contour in Fig. 1d) reveals the upward mass transport in the tropics and the subsequent downward transport in the extratropics of the winter hemisphere in January. This reflects the large-scale Brewer-Dobson circulation in the stratosphere (Butchart, 2014). Tropical injections at 15° N are within the ascending branch of the BD circulation but near the surf zone in the extratropics. A stronger upward transport in the tropics is evident when the volcanic SO₂ injection is further away from the zero line of w*, as shown
- 200 in H1 and H2 in comparison to H5 and H6.

The initial <u>the initial atmospheric</u> conditions control not only the SO_2 and sulfate aerosol transport but also the volcanic halogen transport. More total chlorine and total bromine are transported to NH high latitudes in H6 compared to H2 and H1 (see Section 3.2.3).

The control of initial <u>atmospheric</u> conditions on aerosol and halogen transport and subsequent volcanic forcing is not only evident for tropical eruptions but also for NH extratropical eruptions, although with a different dominating factor. For NH extratropical eruptions at 64° N in January, the initial polar vortex conditions play a dominant role in the transport and evolution as shown in our related paper by Fuglestvedt et al. (2024), the initial stability of the polar vortex strongly influences the lifetime of SO₂ as further investigated in our related paper by Fuglestvedt et al. (2023) and effective radius of SO₄ by controlling the dispersion of injected volcanic gases.

210 3.2 Comparison of tropical and NH extratropical volcanic eruptions with sulfur and halogens injection

3.2.1 SO₂ Sulfur burden and aerosol evolution determines volcanic forcing

Figure 2 shows time series of global and NHET SO_2 , SO_4 , aerosol effective radius (R_{eff}) total sulfur burden and stratospheric aerosol optical depth (SAOD). The global SO_2 burden reduces faster in the first months after tropical eruptions (H-total sulfur burden initially displays a plateau for both tropical (H-15N-Jan, solid lines) compared to NH and extratropical eruptions

215 (H-__64N-__Jan, dashed lines)with an average e-folding time of 2.6 months and 3 months, respectively (Fig. 2a). Along with the reduction of SO₂, SO₄ forms within the first post-eruption month. The maximum global SO₄ burden after tropical and NH extratropical eruptions is almost the same, reaching up to 30 Tg, followed by declines beginning approximately 9 months after the tropical eruptions but 5 months after the and 5 months after tropical eruptions and NH extratropical eruptions, respectively (Fig. 2c). Differences in the transport of SO₂ lead to different peak timing of SO₄ after tropical and NH extratropical eruptions.

- 220 . As is reflected by the NHET SO₂ burden (Fig. 2b), after tropical eruptions (solid lines), SO₂ increases in the first 1 to 2 months and then decreases and disappears 4 to 5 months after the eruption, while showing a gradual transport of SO₂ from the tropics to NH high latitudes. Meanwhile2a). The difference in timing of the start of the decay in sulfur burden between tropical and extratropical eruptions suggests differences in transport processes. Specifically, the NHET SO₄ burden total sulfur burden from tropical eruptions exhibits a sharp increase in the first within the initial 4 to 5 months, followed by a decelerated increase,
- ultimately peaking around at 12 months after the tropical eruptions (Fig. 2d). After the NH extratropical eruptions (dashed lines), due to confinement of the aerosols-14 months (solid lines, Fig. 2b). Conversely, a continuous decline is visible due to aerosol confinement in the NHET region, global and NHET SO₂ and SO₄ burden show similar evolution (Fig. 2a-2d). The duration of global SO₄ burden is affected by the transport of SO₂ from tropical to NH extratropical regions, which is longer for tropical eruptions over 54 months in contrast to NH extratropical eruptions over 36 months. following NH extratropical 230 eruptions (dashed lines, Fig. 2b).

As shown in Figure 1, the initial <u>atmospheric</u> conditions control the transport of volcanic volatiles. The poleward transport of SO_2 and evolution of SO_4 in the NHET (Fig. 2b and 2d) show materials. As a result, the progression of total sulfur burden exhibits a large spread among the six ensemble members of tropical eruptions . More SO_2 is transported to NHET in H6 than in H1, leading to a higher peak NHET SO_4 burden in H6 than in H1. Meanwhile, the global SO_4 burden shows a faster reduction

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a faster reduction of global SO₄ burden (Fig. 2c).

The aerosol size grows through condensation and coagulation along the conversion of SO₂ to SO₄ in the stratosphere. In line with the conversion of SO₂ to SO₄, the global-mean SO₄-mass-weighted mean effective radius (R_{eff} , Fig. 2e) increases faster after tropical eruptions compared to NH extratropical eruptions for the first 6 months . R_{eff} further increases 7–9 months

from its maximum in H6 than in H1 (Fig. 2c). In general, increased NHET transport after tropical eruptions is accompanied by

- 240 after tropical eruptions when the peak has already been reached after NH extratropical eruptions, and it decreases slower after tropical eruptions compared to NH extratropical eruptions. This is similar for the NHET – mean R_{eff} (Fig. 2f) with a smaller but later peak after tropical eruptions compared to NH extratropical eruptions. This agrees with the varied lifetime of SO₂ and SO₄ burden (Fig. 2a–2d). For tropical eruptions, it takes time for SO₂ to be transported northward, and the amount of SO₂ transported to the NHET region varies due to different initial conditions (Fig. 2b). This is also reflected by different R_{eff} in
- 245 different ensemble members. After tropical cruptions, 2). Four months after the eruption, the NHET –mean R_{eff} is larger in maximum, i.e., 0.43 μ m in H6 compared to 0.38 and 0.39 μ m in total sulfur burden peaks at 1.8 Tg and 4.8 Tg in H1 and H2, and decreases faster in H6, respectively. Consequently, 8-10 months later, the peak NHET total sulfur burden is 79% higher in H6 (5.2 Tg) than in H1 and H2 (Fig(2.9 Tg) (Fig. 2b). 2f), indicating a faster removal of larger aerosols with larger gravitational settling velocity than for small aerosols. In line with this, less SO₄ is available outside of NHET regions. Thus, the maximum
- 250 global mean R_{eff} is 0.31 μ m in At the same time, the global total sulfur burden exhibits a 42% longer e-folding time in H1 compared to H6, smaller than 0.36 μ m in H1 and 0.37 μ m in H2 with respective e-folding times of 27 and 19 months (Fig. 2e). For NH extratropical eruptions, the aerosol size grows continuously with SO₂ concentrated in the NHET region. 2a).

 SO_2 , SO_4 , R_{eff} , SO_4 -mass-weighted mean effective radius (R_{eff}) and SAOD evolution are tightly connected and modulated by initial conditions, atmospheric conditions (Figs. 2 and S3, see Section S1). The volcanic forcing is primarily determined by

- 255 the SO₄ burdenwhen the aerosol size exceeds the peak scattering efficiency at 0.25 μm (Lacis, 2015)closely related to the sulfur burden, as shown by a consistent similar evolution of the global-mean and NHET-mean SAOD (Fig. 2g and 2h) and SO₄ (Fig. S3), although fluctuations occur due to variations in effective radius (Fig. S3) with varying scattering efficiency (Lacis, 2015). The maximum global-mean SAOD shows a similar magnitude (~0.2) after tropical and NH extratropical eruptions, but it takes longer to reach the maximum and the forcing lasts longer after tropical eruptions compared to NH extratropical eruptions -
- 260 (Fig. 2c). Due to the wider spread of aerosols, tropical eruptions result in a smaller and delayed peak of NHET-mean SAOD, but a longer-lasting forcing compared to NH extratropical eruptions, after which aerosols are confined in the NHET region -(Fig. 2d).

3.2.2 Climate impact

Figure 3 shows the surface temperature response in the ensembles of tropical and NH extratropical eruptions. Both global-mean

- 265 (Global-mean surface temperature shows a maximum cooling of 0.7 K within 23 months, returning to the control run's 2σ -variability range 42 months after tropical eruptions (solid black line in Fig. 3aand-). Conversely, a maximum cooling of 0.8 K occurs 9 months after extratropical eruptions (dashed black line in Fig. 3c)and-, with temperatures returning to the 2σ -variability range within 24 months. NHET-mean surface temperature (Fig. 3b and 3d) show a weaker but longer-lasting surface cooling displays a maximum cooling of 2.0 K within 22 months, returning to the 2σ -variability range 37 months
- 270 after tropical eruptions (Fig. 3a and solid black line in Fig. 3b)compared to NH. In contrast, a maximum cooling of 2.6 K occurs within 9 months and recovers within 25 months after extratropical eruptions (Fig. 3c and dashed black line in Fig. 3d). The strongest surface cooling emerges in the second and first year after tropical and NH extratropical eruptions, respectively. Extratropical eruptions lead to only slightly stronger global-mean cooling, but much more pronounced NHET-mean surface cooling than tropical eruptions. However, both the global-mean and NHET-mean surface temperatures show a longer-lasting
- 275 cooling after tropical eruptions compared to extratropical eruptions."

The season when the volcano erupts eruption season plays an important role in the temperature variation. When our simulated volcanoes erupt in January, both global-mean and NHET-mean SAOD peak around 12 months after tropical eruptions (Fig. 2g and 2h2c and 2d). During boreal winter, less incoming solar radiation reaches the NH. This interrupts, suspending the reduction of net radiation at the top of the atmosphere (TOA) (Fig. S1a and S1bS4a and S4b) and surface temperature; thus, . Consequently, the cooling peaks in the second year after tropical eruptions (Fig. 3a and 3b). Both global-mean and NHET-mean SAOD peak 5 months after NH extratropical eruptions (Fig. 2g and 2h). This occurs during the boreal summer when the NH experiences its highest solar radiation influx. This synchronization between the largest volcanic forcing and the largest incoming solar radiation leads to the largest reflection of solar radiation (not shown) and subsequent decrease of TOA net radiation (Fig. S1e and S1dS4c and S4d), thus result in the strongest surface cooling in the first year (Fig. 3c and 3d).

285 The surface cooling persists longer after tropical eruptions than NH extratropical eruptions, with variations among the members. Different surface temperature responses in different ensemble members are related to the transport of volcanic volatiles under different initial <u>atmospheric</u> conditions. This is pronounced in the NHET–mean surface temperature responses after tropical eruptions (Fig. 3b). The NHET surface cooling is stronger 5 to 12 months after the eruption in H6 compared

to H1, reflecting the distribution of SO₄ into the NH (Fig. 2d). In addition, the initial ENSO state impacts the temperature

290 response, as reflected by positive global-mean surface temperature anomalies in the first two post-eruption months in H1 and H2 (Fig. 3a and 3c) under initial El Niño states (Table 1) for both tropical and extratropical eruptions.

3.2.3 Halogen–ozone impact

Figure 4 shows the evolution of the total inorganic halogen burden and the ozone response. Injected HCl and HBr in (timeseries in Fig. S5) in the stratosphere reacts with OH to produce chlorine and bromine radicals, reacting with ozone in a catalytic 295 destruction cycle (Solomon, 1999). The global total inorganic chlorine burden shows an e-folding time of 48 and 18 months after tropical and NH extratropical eruptions, including a sizeable plateau at the beginning (Cl_u , Fig. 4a). Global total inorganic bromine burden (Br_u , Fig. 4c) increases for approximately 14 and 6 months after tropical and NH extratropical eruptions, then decreases with e-folding times of 54 and 22 months, respectively. NHET Cl_y and Br_y burdens (solid lines in Fig. 4b and 4d) increase for approximately 14 months, then followed by a continuous decrease with a slight seasonal variation, indicating the 300 transport of halogens into the NHET after tropical eruptions. Halogens are concentrated in the NHET after NH extratropical eruptions, as NHET Cl_y and Br_y burdens (dashed lines in Fig. 4b and 4d) decrease immediately and continuously except for slight variations during the first months. Different ensemble members of tropical eruptions show large differences in the magnitude of NHET Cl_u and Br_u burden, with a larger peak in H6 than in H1 and H2. This indicates that the control of initial atmospheric conditions on SO₂ and aerosol transport is also representative present for halogen transport. The longer transport 305 time to NH high latitudes after tropical eruptions also contributes to a longer lifetime of both global and NHET Cl_{y} and Br_{y} burden, compared to NH extratropical eruptions (Fig. 4a-4d).

In response to the eruptions with co-injection of sulfur and halogens, global-mean and NHET-mean column ozone decreases (Fig. 4e and 4f). Compared to NH extratropical eruptions, tropical eruptions lead to a similar maximum but a delayed peak and longer-lasting global-mean ozone depletion, which lasts over 5 years (Fig. 4e). Due to confinement of halogens

- 310 in the NHET region, NH extratropical eruptions lead to a stronger NHET-mean ozone depletion than tropical eruptions, but the ozone depletion lasts longer after tropical eruptions compared to NH extratropical eruptions. Notably, the NHET-mean column ozone drops below 220 DU, projecting NH ozone hole risks after both tropical and NH extratropical eruptions (Fig. 4f). A larger NHET halogen burden (Fig. 4b and 4d) leads to a stronger NHET-mean ozone depletion in H6 than in H1 and H2 after tropical eruptions. Enhanced northward transport of halogens leads to faster removal as well as less availability of
- 315 halogens in the tropics and Southern Hemisphere. Thus, global-mean halogen burden decreases faster in H6 than in H1 and H2 (Fig. 4a-4d). This leads to earlier depletion and recovery of ozone over both the globe and NHET in H6 than in H1 and H2 (Fig. 4e and 4f).

4 Discussion

4.1 Control of initial atmospheric conditions on the transport of volcanic volatiles

- We interpret the control of the initial <u>atmospheric</u> conditions on the transport of volcanic volatiles after tropical eruptions to be related to the secondary meridional circulation associated with the QBO (Baldwin et al., 2001) and the "leaky pipe" concept of stratospheric circulation in the tropics (Neu and Plumb, 1999). Figure 1 illustrates that H1 and H2 reveal exhibit enhanced upward transport of SO₂, whereas as indicated by a small ratio of poleward/upward transport in contrast to H5 and H6 reveal less upward but more meridional transport towards NH high latitudes (Fig. 1 with a larger poleward transport (a large
- 325 <u>poleward/upward ratio</u>). This stronger leak of SO₂ out of the tropical pipe (for H5 and H6 in contrast to H1 and H2) relates to a stronger stratospheric secondary meridional circulation under a more intense QBO westerly phase (Fig. S2) connected with a larger temperature contrast between tropical and NH mid–latitudes (Fig. S3<u>S6</u>). As pointed out by Ribera et al. (2004), during a strong QBO westerly phase, enhanced ascent of air in the stratosphere is associated with a cooling anomaly in the equatorial area between 15° S and 15° N, while an opposite warming anomaly in the mid–latitudes between 15° N and 55° N
- 330 relates to an enhanced adiabatic sinking. This is reflected in our model results following tropical eruptions (Fig. 1d and S3). S6). In this study, we conducted experiments with a QBO westerly phase at 30 hPa, accompanied with an easterly phase above (Fig. S7). The differences among the ensemble members, particularly regarding the asymmetry of volcanic forcing, might be be amplified if varying initial QBO states are considered, such as including a westerly shear.

The control of initial <u>atmospheric</u> conditions on the transport of volcanic volatiles is also evident after NH extratropical eruptions, in this case primarily controlled by the initial polar vortex state. The northward transport and lifetime of SO₂ increases from members 1 to 6 (Fuglestvedt et al., 2023) (Fuglestvedt et al., 2024) with increasing initial polar vortex stability. Notably, the order of the ensemble members differs between tropical and NH extratropical eruptions, underlining different factors controlling the transport of volcanic volatiles following tropical and NH extratropical eruptions. However, in both this study and Fuglestvedt et al. (2023)Fuglestvedt et al. (2024), the ensembles exhibit less northern poleward transport of SO₂ in

- 340 the first to fourth members, as compared to the fifth and sixth members. The former four members (H1 to H4) correspond to El and H2 correspond to initial El Niño or neutral ENSO initial states, while the states, while H3 and H4 correspond to neutral ENSO states. The latter two members(, H5 and H6), correspond to La Niña states (Table 1). The labelling of the ensemble members, based on the first month's SO2 transport, may correlate with the ENSO state and would also provide insights into the influence of ENSO on the polar vortex. As highlighted in van Loon and Labitzke (1987), El Niño events are typically linked
- 345 to a warming and weakening of the polar vortex, along with a cooling in the tropical lower stratosphere, however, unusual warming occurs when. Nevertheless, a simultaneous injection of volcanic gases and aerosols takes placecan counteract this ENSO-related cooling, resulting in unusual tropical stratospheric warming. In contrast, La Niña events often demonstrate are often connected with a strengthening of the polar vortex. Future studies with large ensemble simulations should be conducted to investigate the ENSO-polar vortex-volcanic eruption connection.
- 350 Overall, we interpret our results as being primarily influenced by initial conditions such as the state of show that initial atmospheric conditions impact the distribution of volcanic materials already in the first month as well as the following evolution

and distribution of volcanic aerosols, ultimately shaping the QBO, ENSO, and the stratospheric polar vortex. These factors collectively control the transport of volcanic volatiles and aerosolsand ultimately determine the volcanic forcing. Certainly, ambient meteorological conditions introduce an element of unpredictability to the dispersion of volcanic materials out of

- 355 the tropics. However, as highlighted by Jones et al. (2016), this dispersion is particularly sensitive to ambient meteorological conditions for low-altitude eruptions (16–18 km). In contrast, a high-altitude eruption scenario (19–29 km) exhibits reduced meteorological variability, a phenomenon to which our simulations, with injection altitude of 24 km, appear to be less susceptible. Our results reveal the potential for predicting the latitudinal distribution of volcanic volatiles and assessing the subsequent volcanic forcing and associated climate and environmental impacts already after the first month of tropical and NH extra-
- 360 tropical eruptions. Other models and ensemble members are necessary to validate More models and large ensembles are needed to corroborate these findings, and to test different initial states with the QBO easterly phase. Zanchettin et al. (2022) recommended improving the assessment of initial condition influences on direct radiative and dynamical responses how different initial climate states such as ENSO and the QBO could impact the distribution of volcanic materials, volcanic forcing and subsequent responses for tropical and extratropical eruptions. The future phase of the VolMIP-Model Intercomparison 365 Project on the climatic response to Volcanic forcing (VolMIP) (Zanchettin et al., 2022) holds potential as a valuable resource
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4.2 Comparisons between tropical and NH extratropical eruptions

for addressing these research needs.

Based on tree ring proxy data and MAECHAM5–HAM model simulations, Toohey et al. (2019) concluded that compared to NH extratropical eruptions, tropical eruptions lead to weaker maximum volcanic forcing over the NHET for a given injection
height with sulfur–only injection. Figure 5 presents an overall summary of the comparison between tropical and NH extratropical eruptions regarding their maximum volcanic forcing and impacts on surface temperature and ozone. Our fully coupled model results confirm the results from Toohey et al. (2019), also when co–injecting sulfur and halogens and resolving interactive atmospheric chemistry for the simulated volcanic eruptions. However, when considering the time–to–maximum and the lifetime, tropical eruptions cause longer–lasting volcanic forcing and surface cooling compared to NH extratropical eruptions, over both the globe and NHET. The delayed response time after tropical eruptions results from the northward transport of stratospheric aerosols (Fig. 2). Sulfur was the only injected volcanic volatile and OH was prescribed in MAECHAM5–HAM. Thus, the timing difference is lacking for eruptions at different latitudes in Toohey et al. (2019). Including interactive atmospheric chemistry and OH in models can be important in order to simulate the timing of volcanic aerosol evolution, volcanic forcing and subsequent impact, as previously also noted by Mills et al. (2017) and Clyne et al. (2021).

380 4.3 Sensitivity to varying eruption source parameters

Here we discuss how sensitive our results are to varying eruption source parameters. Figure 6 shows the global-mean and NHET-mean SAOD, surface temperature and column ozone variations after tropical and NH extratropical eruptions with different volcanic volatile injections and in different seasons. We focus on testing sulfur-only injections and eruptions in July (see Table 1). The simulated maximum global-mean SAOD is larger-5% higher in H–15N–Jan than in S–15N–Jan, and even

- $\frac{10\% \text{ higher in H-15N-Jul than in S-15N-Jul Besides, both S-15N-Jan and S-15N-Jul fall outside the H-15N-Jan}{\pm 2\sigma \text{ range (Fig. 6a)}}$ $\frac{10\% \text{ higher in H-15N-Jul than in S-15N-Jul Besides, both S-15N-Jan and S-15N-Jul fall outside the H-15N-Jan}{\pm 2\sigma \text{ range (Fig. 6a)}}$ $\frac{10\% \text{ higher in H-15N-Jul than in S-15N-Jul Besides, both S-15N-Jan and S-15N-Jul fall outside the H-15N-Jan}{\pm 2\sigma \text{ range (Fig. 6a)}}$ $\frac{10\% \text{ higher in H-15N-Jul than in S-15N-Jul Besides, both S-15N-Jan and S-15N-Jul fall outside the H-15N-Jan}{\pm 2\sigma \text{ range (Fig. 6a)}}$ $\frac{10\% \text{ higher in H-15N-Jul than in S-15N-Jul Besides, both S-15N-Jan and S-15N-Jul fall outside the H-15N-Jan}{\pm 2\sigma \text{ range (Fig. 6a)}}$ $\frac{10\% \text{ higher peak global-mean SAOD after tropical eruptions in Julywith co-injection of The percentage change in July eruptions closely aligns with the findings of Staunton-Sykes et al. (2021) for a 10 Tg SO2 injection in July, revealing a 11\% higher peak global-mean SAOD when sulfur and halogens than sulfur-only were co-injected compared to sulfur-only were co-injected compared t$
- injection. However, this relationship is also dependent on the eruption latitude. After NH extratropical eruptions, the difference of global-mean SAOD is limited and differs after eruptions in different seasons (Fig. 6c). The maximum NHET-mean SAOD (Fig. 6b and 6d) and surface temperature response (Fig. 6e-6h) also varies among different experiments with varying volcanic volatiles as well as eruption latitudes and eruption seasons. Nonetheless, these responses are mostly within the range of H-15N-Jan ±2σ and H-64N-Jan ±2σ, indicating a potential larger impact of varying initial atmospheric conditions than
 tested varying eruption source parameters on the volcanic forcing and subsequent surface cooling.

Varying volcanic volatiles, particularly with or without halogen injection, has a significant impact on stratospheric ozone under pre–industrial conditions. In line with Brenna et al. (2020), volcanic eruptions with co–injection of sulfur and halogens result in substantial ozone depletion (solid lines), while volcanic eruptions with sulfur–only injection (dashed lines) slightly increase global and NHET ozone levels (Fig. 6i and 6j). Co–injection of sulfur and halogens leads to similar maximum ozone

- 400 depletion after tropical and NH extratropical eruptions in January (H–15N–Jan and H–64N–Jan), but a stronger ozone depletion occurs after the tropical eruption compared to the NH extratropical eruption in July (H–15N–Jul and H–64N–Jul). For eruptions at the same latitude, the maximum ozone loss is larger in H–15N–Jul than in H–15N–Jan, but smaller in H–64N–Jul than in H–64N–Jan. The increase of ozone following sulfur–only injections is stronger after tropical eruptions compared to NH extratropical eruptions. Tropical sulfur–only eruptions lead to significant ozone increases (S–15N–Jan and S–15N–Jul),
- 405 while ozone responses after NH extratropical eruptions (S-64N-Jan and S-64N-Jul) mostly fall within the range of two standard deviations of the control run. The impact of tested varying source parameters is generally smaller than the impact of varying initial <u>atmospheric</u> conditions, as the sensitivity experiments mostly fall within the modelled range of H-15N-Jan $\pm 2\sigma$ and H-64N-Jan $\pm 2\sigma$. Further studies with ensembles for the sensitivity tests are needed to further clarify the role of initial <u>atmospheric</u> conditions and varying eruption source parameters in affecting volcanic forcing and subsequent climate and
- 410 environmental impacts.

In this study, we only considered focused on the co–injection of sulfur and halogens . Zhu et al. (2020) and Stenchikov et al. (2021) have shown that co–injection of sulfur, ash and water vapor affect in our baseline experiments and conducted sensitivity tests to examine the impact of sulfur-only injection. Volcanic ash and other volatiles, such as water vapor, can also be injected (LeGrande et al., 2016; Millán et al., 2022; Zhu et al., 2020), which can alter the composition, distribution and radiative forcing

415 of a volcanic cloud during the first monthsvolcanic forcing. However, whether these volcanic materials influence the maximum, time-to-maximum and lifetime of volcanic forcing in combination with sulfur and halogen injections has not been studied yet. Future simulations that incorporate these volcanic materials are needed to investigate how they affect the results shown in this study.

4.4 Model dependency

- 420 Clyne et al. (2021), Marshall et al. (2018) and Quaglia et al. (2023) showed that the impacts of varying source parameters on volcanic forcing and related climate responses are model dependent. Here, we discuss our model results on sulfur-only injection experiments with MAECHAM5–HAM and EVA (Fig. 6). After tropical eruptions, similar maximum global-mean SAOD emerges after January and July eruptions in MAECHAM5–HAM (green lines), but peaks at least 6 months earlier than in CESM2–WACCM6 (red dashed lines, Fig. 6a). This may relate to the interactive OH in CESM2–WACCM6 (Fig.
- 425 \$4\$\$8). Maximum NHET-mean SAOD in MAECHAM5-HAM is smaller compared to CESM2-WACCM6 (Fig. 6b). For NH extratropical eruptions, maximum global and NHET-mean SAOD is much smaller after the July eruption compared to the January eruption in MAECHAM5-HAM, but limited differences are shown in CESM2-WACCM6 (Fig. 6c and 6d). In comparison, EVA (gray lines) generally produces smaller SAOD compared to CESM2-WACCM6 and MAECHAM5-HAM, except for that in MAECHAM5-HAM-64N-Jul (Fig. 6b). The inter-model difference shown in this study agrees with Clyne
- 430 et al. (2021), where CESM–WACCM showed the largest maximum global–mean, the longest time–to–maximum, and the longest duration of the SAOD. Notably, CESM–WACCM produced the smallest global mean stratospheric effective radius among the models. For Pinatubo–strength simulations, our CESM2–WACCM6 results also indicate a smaller global–mean R_{eff} (=0.35 μ m) compared to a R_{eff} exceeding 0.4 μ m with MAECHAM5–HAM (Toohey et al., 2019)). These differences can arise from different model top, aerosol microphysics or chemistry resolved in model configurations. Zanchettin et al. (2022)
- 435 showed that, under the VolMIP protocol, the maximum global-mean surface cooling ranges between 0.27 °C and 0.38 °C in ensemble means of six models simulating the Pinatubo eruption, which is lower than the analyzed 0.5 °C cooling based on HadCRUT5 observational data. In comparison, our CESM2–WACCM6 runs simulate a global-mean maximum cooling up to 0.6 °C in S–15N–Jul. In line with Zanchettin et al. (2016), CESM2–WACCM6 represents an upper threshold among the models.

440 **5** Summary and conclusions

In this study, we perform idealized experiments with CESM2–WACCM6 to compare tropical and Northern Hemisphere extratropical volcanic eruptions in terms of the evolution of volcanic volatiles and aerosols, volcanic forcing and subsequent impacts on surface temperature and ozone. We consider in particular the co–injection of sulfur and halogens to the stratosphere under pre–industrial 1850s conditions and investigate the sensitivity to initial <u>atmospheric</u> conditions. We ran two sets of baseline experiments with volcanic eruptions in January at 15° N and 64° N co–injecting SO₂ and halogens (HCl, HBr) into the stratosphere at 24 km altitude. Each set has six ensemble members with westerly OBO and varying ENSO and polar vortex initial

states. We also performed sensitivity tests varying volcanic volatiles (sulfur only) and eruption season (July).

Our model results reveal that initial <u>atmospheric</u> conditions control the meridional transport of sulfur and halogens in the first month after the eruptions, and further modulate the latitudinal distribution of sulfate aerosols, halogens, volcanic forcing and impacts. The baseline experiments, with the same eruption source parameters but different initial <u>atmospheric</u> conditions,

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show large variations in the volcanic forcing and subsequent climate and ozone impacts. For tropical eruptions, the initial atmospheric conditions also control the hemispheric asymmetry of the volcanic forcing.

Tropical and NH extratropical volcanic eruptions co–injecting SO₂ and halogens lead to similar maximum global-mean aerosol forcing, surface cooling and ozone depletion. However, for NHET-mean responses, the NH extratropical eruptions have a larger maximum impact than tropical eruptions. For both global-mean and NHET-mean volcanic forcing, climate and ozone impacts, tropical eruptions take longer to peak and last longer than NH extratropical eruptions. Thus, when evaluating whether tropical or NH extratropical eruptions have larger climate and environmental impacts, it is important to evaluate both the severity and the duration of the impact, as different perspectives can come to different conclusions.

Compared to volcanic eruptions with sulfur–only injection, co–injection of sulfur and halogens leads to large differences in 460 the ozone responses. Under pre–industrial conditions, volcanic eruptions with sulfur–only injections lead to a slight increase of ozone. In contrast, co–injection of sulfur and halogens leads to a significant ozone depletion lasting more than 5 years after tropical eruptions and up to 5 years after NH extratropical eruptions of Pinatubo–strength.

Compared to varying the source parameters of volcanic eruptions, varying initial conditions reveal the initial atmospheric conditions reveals an important impact on the volcanic forcing and subsequent climate and ozone impacts. The impact has a

- 465 larger variability at the early stage after tropical eruptions than after NH extratropical eruptions, as initial <u>atmospheric</u> conditions lead to large differences in the transport of volcanic volatiles and aerosols from tropics to high latitudes already from the first post-eruption month. For NH extratropical eruptions, the variability of surface temperature response increases over time, especially during winter.
- Both CESM2–WACCM6 and MAECHAM5–HAM model results show varied global–mean and NHET–mean SAOD with eruptions at different latitudes and in different seasons. Compared to both models, EVA produces similar forcing duration but smaller global–mean and NHET–mean maximum SAOD, and with limited variations to different eruption latitudes and eruption seasons. The differences in volcanic forcing revealed here underscores underscore a potential large discrepancy between utilizing Earth system models with interactive atmospheric chemistry and models with prescribed SAOD generated with EVA to examine the influence of varying initial <u>atmospheric</u> conditions and source parameters on volcanic forcing and subsequent
- 475 impacts.

Initial conditions and eruption source parameters seem to be key for understanding the difference and comparability between simulations and observations of forcing and impacts of volcanic eruptions. Our study highlights the large variability of volcanic forcing and response with varying initial <u>atmospheric</u> conditions, which is comparable to that with varying eruption source parameters. Multi–model simulations and comparisons are needed to further test our results. Our results point to the

480 necessity of including initial conditions in future VolMIP protocols for better understanding volcanic impacts on the climate and environment.

Data availability. The CESM2–WACCM6 data generated and analysed for the current study are available in the NIRD Research Data Archive.

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Author contributions. Z.Z., H.F., M.T. and K.K. designed the model experiments and Z.Z., H.F. and M.T. carried them out. Z.Z. prepared the manuscript with contributions from all co-authors. K.K. led the project.

Competing interests. One of the (co-)authors is a member of the editorial board of Atmospheric Chemistry and Physics.

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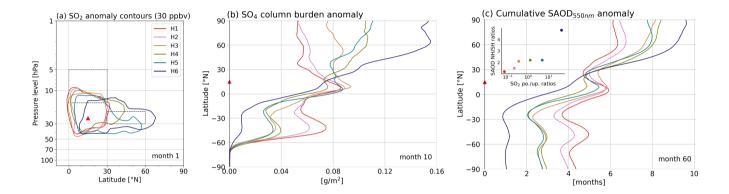


Figure 1. Latitudinal First month latitudinal and vertical distribution of the first month SO₂ column burden concentration anomaly at 30 ppby (contour, a), the sixth- 10^{th} month SO₄ column burden anomaly (b) , and 60 months cumulative SAOD anomaly at 550 nm (c) , and the first month latitudinal and vertical distribution of SO₂ concentration anomaly (shades), and residual vertical velocity (w*, contours with an interval of 0.5 mm s⁻¹) (d) in the stratosphere after tropical eruptions in January for six members of the baseline experiments experiment. The ensemble members are ordered in an ascending trend of northward SO₂ transport in the first month. The red triangle indicates denotes the eruption latitude at 15° N in panels (a-c), and also the injection altitude at 24 km in (da). The inset axis in (c) indicates the relationship between the poleward/upward ratio of the first month SO₂ concentration anomaly and the NH/SH ratio of the 60 months cumulative SAOD anomaly.

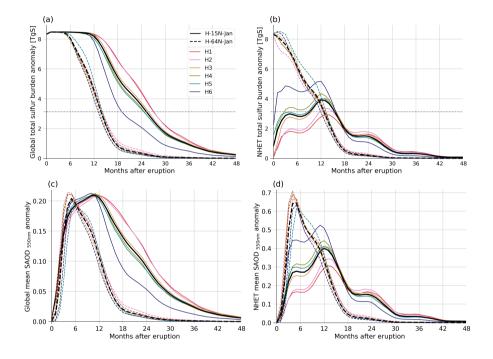


Figure 2. Global (left panel) and Northern Hemisphere extratropics (NHET $[30 \degree N - 90 \degree N]$, right panel) SO₂-total sulfur burden (a, b) , SO₄ (c, d) burden anomaly, aerosol effective radius (R_{eff} , e, f) and stratospheric aerosol optical depth (SAOD, gc, hd). Solid lines and dashed lines are for tropical and NH extratropical eruptions, respectively. Different colors represent different ensemble member realizations. The black lines represent the ensemble means of the baseline experiments. The gray dotted horizontal line in (a) represents the e-folding of injected sulfur. Note the different axes in different subfigures(c) and (d).

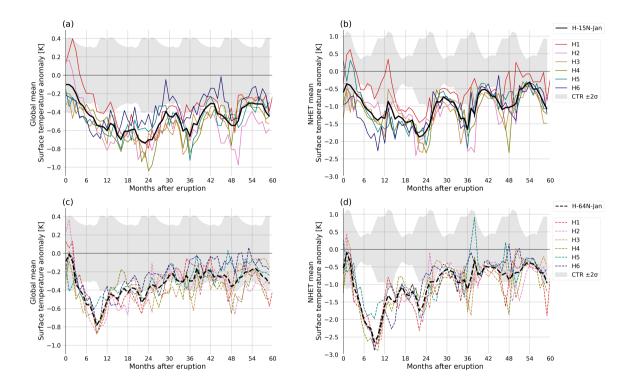


Figure 3. Global (left panel) and Northern Hemisphere extratropics (NHET, right panel) mean surface temperature anomaly. Solid lines and dashed lines are for tropical (a–b) and NH extratropical (c–d) eruptions, respectively. Different colors represent different ensemble member realizations. The black lines represent the ensemble means of the baseline experiments. The gray shades represent two standard deviations of the control run. Note the different y–axes between the left and right panels.

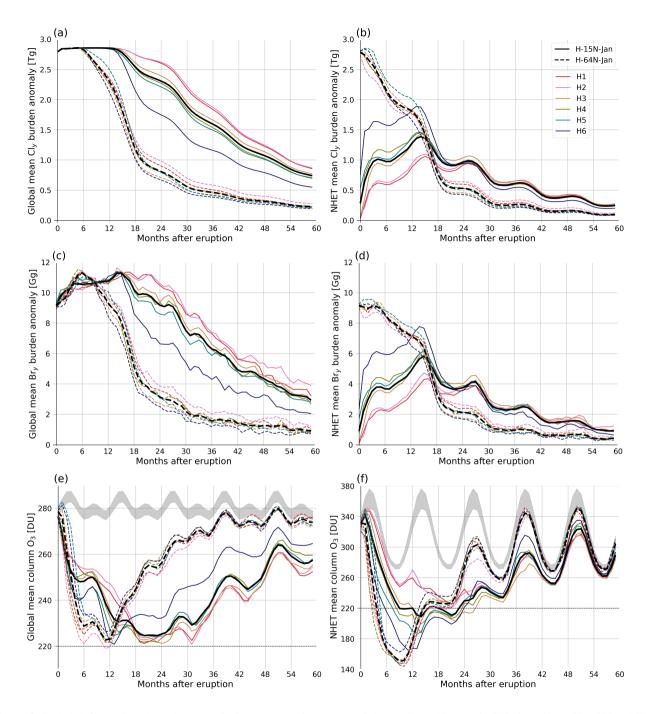


Figure 4. Global (left panel) and Northern Hemisphere extratropics (NHET, right panel) total inorganic Chlorine ($Cl_y = Cl + ClO + 2Cl_2 + 2Cl_2O_2 + OClO + HOCl + ClONO_2 + HCl + BrCl, a, b$) and total inorganic bromine ($Br_y = Br + BrO + HOBr + BrONO_2 + HBr + BrCl, c, d$) burden anomaly and global and NHET mean column O₃ (e, f). Solid lines are variations after tropical eruptions, while dashed lines are variations after NH extratropical eruptions. Different colors represent different ensemble member realizations. The black lines represent the ensemble means of the baseline experiments. The gray dotted horizontal line in (a) and (c) represents the e-folding of injected halogens. The gray shades (e, f) represent two standard deviations of the control run.

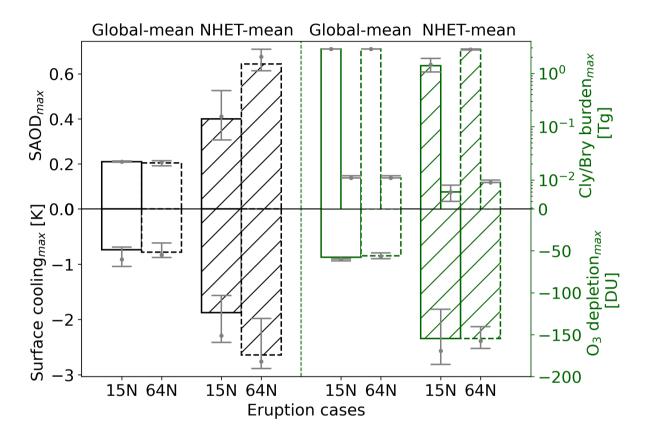


Figure 5. Summary of global–mean and NHET–mean maximum volcanic forcing (left top part), surface cooling (left bottom part), halogen burden (right top part) and ozone depletion (right bottom part) between tropical and NH extratropical volcanic eruptions at 15° N (solid bars) and 64° N (dashed bars), respectively. In the top right panel, the high and low bars are Cl_u and Br_u , respectively. The grey error bars and grey dots indicate the range and median of the six ensemble members of the baseline experiments.

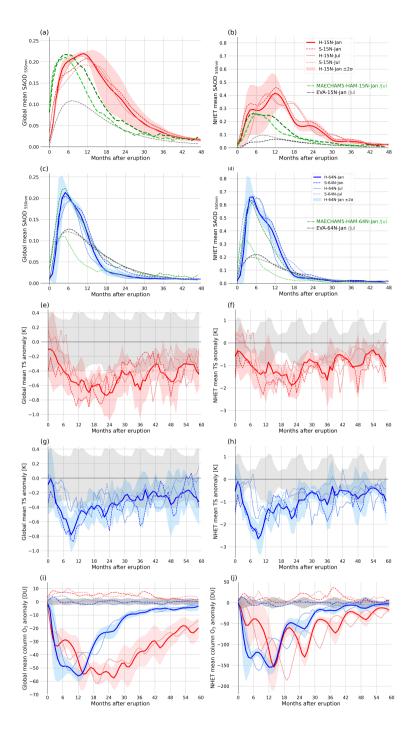


Figure 6. Global (left panel) and Northern Hemisphere extratropics (NHET, right panel) mean stratospheric aerosol optical depth (SAOD) at 550 nm (a–d), surface temperature anomaly (e–h) and column ozone anomaly (i, j) after tropical and NH extratropical eruptions. The thick lines and the shades represent two standard deviations of are for the baseline ensemble experimentsmean and two-standard deviation of the six members-baseline experiment. The thin lines are for sensitivity tests. Red and blue color lines and shades colors represent variations after tropical and NH extratropical eruptions, respectively. The solid and dashed lines are simulated volcanic eruptions with co–injection of sulfur and halogen and sulfur–only injection, respectively. The gray **28**ades (e–j) represent two standard deviations of the control run. Note the different y–axes between the left and right panels.

CAVA *: 15° N, 91° W Iceland: 64° N, 19° W H-15N-Jan # H-64N-Jan # S-15N-Jan # H-64N-Jan # S-15N-Jul S-64N-Jul S-15N-Jul S-64N-Jul	member H1 H2 H3 H4 H5 H6	@ 30 hPa Westerly Westerly Westerly	(INO)				
H-64N S-641 H-641	H1 H2 H3 H4 H5 H6	Westerly Westerly Westerly		+	SO ₂ [Tg]	HCl* [Tg]	HBr * [Gg]
	H2 H3 H4 H5 H6	Westerly Westerly Westerly	El Niño	3	17	2.93	9.5
	H3 H4 H5 H6	Westerly Westerly	El Niño	1			
	H4 H5 H6	Westerly	Neutral	4			
	H5 H6	ATTOICOTT &	Neutral	2			
	9H	Westerly	La Niña	9			
		Westerly	La Niña	5			
		Westerly	neutral	4	17		
		Westerly	neutral		17	2.93	9.5
		Westerly	neutral		17		
ECHAM5–15N–Jan # ECHAM5–64N–Jan				I	17	1	
ECHAM5–15N–Jul # ECHAM5–64N–Jul					17		
EVA-15N-Jan EVA-64N-Jan		I		I	17	1	I
EVA-15N-Jul EVA-64N-Jul		I	I		17		

Table 1. Summary of model experiments with CESM2-WACCM6. In addition, MAECHAM5-HAM (ECHAM5) and EVA as stated.

stratosphere (Krüger et al., 2015; Brenna et al., 2019).

†Numbers of initial polar vortex (PV) states as in Fuglestvedt et al. (2024).

Ensemble mean.