

Improved Simulation of Volcanic Sulfur Dioxide Depletion and Transport Using a High-Resolution Volcanic Sulfur Dioxide Emissions Inventory

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Abstract

Explosive volcanic eruptions inject substantial amounts of sulfur dioxide (SO_2) into the upper troposphere and stratosphere, forming sulfate aerosols that persist for months to years and perturb Earth's radiative and chemical environment. Accurately simulating these impacts requires accurate emission timing and injection altitude, which remain only coarsely represented in conventional volcanic SO_2 inventories. Here, we present a high-resolution Time- and Height-Resolved Inventory of Volcanic Emissions of SO_2 (THRIVE- SO_2) that explicitly resolves the temporal evolution and vertical distribution of SO_2 emissions for 52 explosive volcanic eruptions since 2002. We evaluate its performance against the VolcanEESM global volcanic SO_2 emissions database using five representative eruptions.

Compared with the VolcanEESM database, THRIVE- SO_2 more accurately reproduces the observed multi-day evolution of SO_2 emissions and their vertically resolved injection profiles. Simulations driven by THRIVE- SO_2 show closer agreement with satellite observations for nearly all cases: they better reproduce the observed buildup and subsequent depletion of SO_2 mass, as well as the spatiotemporal evolution of SO_2 plumes. The improved representation of emission timing and injection altitude also leads to more realistic sulfate aerosol evolution, including latitude–altitude distributions and long-range transport. In particular, resolving the vertical injection profile reduces the artificial placement of SO_2 in the stratosphere and helps avoid overestimation of stratospheric aerosol loading. These results show that improved simulation of volcanic SO_2 and sulfate aerosols is primarily due to the more realistic temporal evolution and vertical distribution of SO_2 emissions in THRIVE- SO_2 . THRIVE- SO_2 therefore provides a robust emission basis for investigating volcanic SO_2 transport and assessing the atmospheric and climatic impacts of explosive eruptions.

Introduction

Explosive volcanic eruptions can inject substantial amounts of sulfur dioxide (SO_2) into the upper troposphere and stratosphere, where subsequent oxidation converts SO_2 into sulfate aerosol particles. Because of their high stability and limited depositional removal in this region, these aerosols can persist for months to years and undergo long-range, sometimes near-global, transport. Volcanic sulfate aerosols can substantially perturb the Earth's climate system through radiative and chemical effects. They scatter incoming solar radiation, thereby reducing the shortwave radiation reaching the surface and potentially inducing surface cooling, while their radiative absorption can contribute to lower-stratospheric warming. In addition, these aerosols provide surfaces for heterogeneous chemical reactions and may promote stratospheric ozone depletion, with potential consequences for atmospheric circulation and the hydrological cycle. Although no eruptions comparable to Mt. Pinatubo in stratospheric sulfur injection have occurred in the past two decades, recurrent eruptions have measurably perturbed the stratospheric aerosol layer and, collectively, contributed to its interannual variability and radiative forcing^{1–4}.

Accurately quantifying the impacts of volcanic eruptions requires considering not only their total SO₂ emissions but also their initial emission characteristics and the meteorological and chemical environment the volcanic plume encounters. In particular, the emission timing, injection altitude, and vertical mass distribution determine the dynamical pathways and atmospheric conditions governing aerosol formation, transport, and removal^{5–7}. However, existing volcanic SO₂ inventories often have limited temporal and spatial resolution, which can reduce the accuracy of simulations of volcanic SO₂ depletion and aerosol dispersion.

Existing volcanic SO₂ inventories are primarily derived from satellite observations, including the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS)^{8–9}, Ozone Monitoring Instrument (OMI)⁹, the Infrared Atmospheric Sounding Interferometer (IASI), and the Ozone Mapping and Profiling Suite (OMPS)¹⁰. Satellite measurements provide information on SO₂ column amounts and horizontal plume distributions, from which the total SO₂ mass and approximate plume-height range can be estimated. However, limitations in sampling frequency, viewing geometry, and retrieval assumptions hinder direct characterization of continuous, detailed SO₂ vertical and temporal variations in emission rate, injection altitude, and eruption duration. Consequently, existing inventories often cannot fully represent the temporal variability and vertical heterogeneity of volcanic SO₂ emissions.

For example, the VolcanEESM global volcanic SO₂ emissions database¹⁰, the volcanic SO₂ inventory commonly used in the Whole Atmosphere Community Climate Model (WACCM) in the Community Earth System Model (CESM), provides the total SO₂ mass, eruption date, and lower and upper plume altitudes for each event. To meet global model input requirements, emitted SO₂ is typically released into the model grid cell containing or adjacent to the volcano over a fixed 6-h period (12:00–18:00 UTC) on the eruption day and distributed uniformly by volume between the reported lower and upper plume altitudes^{11–12}. This approach assumes a fixed emission duration, constant emission rate, and vertically uniform distribution. However, volcanic eruptions can exhibit pronounced temporal variability and vertical inhomogeneity. The chemical transformation, transport, and removal of SO₂ depend on its injection altitude and the atmospheric environment the plume encounters. Therefore, simplified assumptions about emission timing and vertical allocation may bias simulations of plume evolution and radiative effects¹³. A higher-resolution, altitude- and time-resolved volcanic SO₂ inventory is needed to enable more realistic simulations.

In response to this need, we developed a high-resolution, altitude- and time-resolved volcanic SO₂ emission inventory covering 52 eruptions from 2002 to the present, referred to as the Time- and Height-Resolved Inventory of Volcanic Emissions of SO₂ (THRIVE-SO₂)¹⁴. In contrast to inventories such as VolcanEESM, THRIVE-SO₂ is constructed by combining Atmospheric Infrared Sounder (AIRS) SO₂ observations¹⁵ with backward trajectory calculations: particles are initialized across the observed SO₂ plume and traced backward in time to determine when and at what altitude they passed through the volcanic source region, yielding an altitude- and time-resolved emission profile with a resolution of 0.2

km×15 minutes for each eruption. This trajectory-based approach enables THRIVE-SO₂ to resolve the temporal evolution of emission rate and injection height throughout an eruption, rather than assuming a fixed release duration and vertically uniform distribution. Detailed descriptions of the inventory construction are provided in Supplementary Note 1.

THRIVE-SO₂ has been used to initiate simulations of SO₂ and aerosol transport for several volcanic eruptions, providing critical insights into key transport pathways for volcanic aerosols in the upper troposphere and lower stratosphere (UTLS)^{6–7, 13, 16–18}. For example, THRIVE-SO₂-based simulations revealed the quasi-horizontal transport of Sarychev aerosols into the tropical tropopause layer (TTL) via the clockwise circulation of the South Asian High (SAH)⁶, the diabatic lofting of Nabro aerosols across the tropopause within the SAH⁷, and the rapid quasi-horizontal mixing that transported Merapi aerosols from the tropics to the Antarctic lower stratosphere following the breakdown of the polar vortex¹⁶.

Although THRIVE-SO₂ was originally developed to enable more realistic simulations of volcanic aerosol transport, it would be even more valuable if it could also more accurately reproduce observed SO₂ mass depletion and aerosol loading. Assessing this capability is a critical first step toward evaluating the potential benefits of using this inventory for climate impact assessments. Furthermore, studies of geoengineering via stratospheric sulfur injection show that the amount, geographic location, and injection height of SO₂ emissions strongly control the climatic effects of stratospheric sulfate aerosols^{19–22}, underscoring the value of a more accurate, altitude-resolved SO₂ inventory for improving climate-impact simulations.

The present study evaluates the improvements brought by THRIVE-SO₂ through five representative eruption cases: Sarychev (48.1°N, 153.2°E) in June 2009, Merapi (7.54°S, 110.44°E) in 2010, Nabro (13.4°N, 41.7°E) in June 2011, Kelud (7.93°S, 112.31°E) in 2014, and Calbuco (41.33°S, 72.61°W) in 2015. By comparing volcanic SO₂ and aerosol simulations driven by THRIVE-SO₂ against those driven by the VolcanEESM global volcanic SO₂ emissions database, we assess whether THRIVE-SO₂ yields improvements in reproducing observed SO₂ evolution and aerosol loading.

Results

We selected five cases based on their distinct transport pathways and dynamical contexts.

(1) The 2009 Sarychev eruption serves as a Northern Hemisphere event, in which volcanic aerosols were injected into the mid-latitude lower stratosphere and were transported equatorward into the TTL via the clockwise circulation of the SAH during boreal summer. Once entrained into the TTL, these aerosols gained access to the global stratospheric circulation⁶.

(2) The 2010 Merapi eruption demonstrates how tropical volcanic aerosols can be transported quasi-horizontally from the TTL to the polar lower stratosphere, and subsequently descend into the Antarctic

lower stratosphere following polar vortex breakdown in austral spring, driving significant polar ozone loss and illustrating the efficiency of interhemispheric transport pathways¹⁶.

(3) The 2011 Nabro eruption exemplifies a tropical case where a substantial fraction of SO₂ was injected only into the upper troposphere, yet was subsequently lofted across the tropopause into the stratosphere through the confinement and upward motion within the Northern Hemisphere SAH anticyclone, highlighting how dynamical uplift can compensate for initially low injection heights⁷.

(4) The 2014 Kelud eruption represents a tropical eruption that injected SO₂ into the upper troposphere and stratosphere, with the plume top exceeding 25 km. Despite its moderate magnitude, the injected SO₂ substantially increased aerosol loading in the tropical and Southern Hemisphere lower stratosphere in 2014, producing appreciable radiative forcing. The eruption also emitted substantial volcanic ash, which persisted in the lower stratosphere and complicated the identification and quantification of sulfate aerosol and SO₂ from satellite observations²³.

(5) The 2015 Calbuco eruption represents a Southern Hemisphere mid-latitude event, in which volcanic aerosols were transported poleward into the Antarctic stratosphere, where they contributed to heterogeneous chemistry and stratospheric ozone depletion over Antarctica²⁴.

Together, these five cases span both hemispheres and encompass a broad range of source latitudes, emission altitudes, and dynamical transport regimes, providing a comprehensive testbed for evaluating the THRIVE-SO₂.

Comparison of volcanic SO₂ emission inventories

Figure 1 compares the SO₂ emissions for the five eruptions from THRIVE-SO₂ and VolcanEESM. For each eruption, THRIVE-SO₂ scales the total SO₂ mass to match VolcanEESM. THRIVE-SO₂ is provided with vertical and temporal resolution of 15 minutes × 0.2 km. VolcanEESM assumes that volcanic SO₂ is released for 6 hours (12:00–18:00 UTC) on the eruption day and is uniformly distributed in volume between the reported lower and upper plume altitudes.

Figure 1 highlights substantial differences in both the temporal evolution and vertical distribution of the emissions between these two inventories. THRIVE-SO₂ generally provides a more vertically structured and time-varying representation of the emissions, clearly distinguishing whether SO₂ is injected into the troposphere or the stratosphere.

(a)

Figure 1. SO₂ emission time series and vertical profiles of the Sarychev eruption (2009), Merapi eruption (2010), the Nabro eruption (2011), the Kelud eruption (2014), and the Calbuco eruption (2015). (a)–(e) are SO₂ emissions from THRIVE-SO₂. The bin size is 15 minutes × 0.2 km. Black dots

indicate tropopause height. (f)–(j) are SO₂ emissions from VolcanEESM. Panels (k)–(o) compare the vertical profiles of the SO₂ emission integrated over the eruption period and interpolated onto the model levels of the WACCM model, indicated by the light gray horizontal lines.

The SO₂ emissions from these two inventories, integrated over the eruption period and interpolated onto WACCM model levels, as shown in Fig. 1k–o, provide a more direct, quantitative comparison of differences between the two inventories. Figure 1k–o shows that VolcanEESM generally captures the altitude range over which large volcanic SO₂ emissions occur in the upper troposphere and stratosphere, retaining the portion of emissions most likely to exert substantial climate impacts. THRIVE-SO₂ and VolcanEESM show similar vertical distributions for the Nabro eruption, whereas the other cases exhibit notable differences in both emission magnitude and vertical spread, implying potentially different subsequent transport pathways and radiative–chemical impacts. Consequently, the two inventories may expose SO₂ to different chemical and dynamical environments, leading to differences in both the initial mass evolution and subsequent depletion rate.

Comparison of volcanic SO mass depletion and plume transport

We simulated the volcanic SO₂ depletion using the Lagrangian transport model Massive-Parallel Trajectory Calculations (MPTRAC) Version 2.6²⁵, with the THRIVE-SO₂ and VolcanEESM separately prescribed as the initial SO₂ emissions. Except for the different temporal and spatial distribution of the initial SO₂ emissions, the two sets of simulations were performed with identical model configurations, including the total SO₂ masses, number of particles released, physical and chemical settings, and meteorological driving data, so that differences in the simulated SO₂ evolution can be attributed to the differences in the emission inventories.

Figure 2 compares the time series of total SO₂ mass derived from the Ozone Monitoring Instrument (OMI)²⁶ with MPTRAC simulations initialized using the THRIVE-SO₂ and VolcanEESM. Both simulations broadly reproduce the persistence and post-eruptive decline of the volcanic SO₂ burden, although their agreement with OMI varies among eruptions.

(c)

Figure 2. Comparison of the simulated total SO₂ mass time series initialized by the THRIVE-SO₂ and VolcanEESM against OMI SO₂ observations for each eruption case.

Pronounced discrepancies between the two simulations emerge during the initial eruption stage and persist as the SO₂ mass declines. The THRIVE-SO₂ simulations generally reproduce the progressive increase in SO₂ mass as observed by OMI, whereas the VolcanEESM simulations show an abrupt appearance of the volcanic SO₂ burden and therefore do not resolve the initial buildup. This discrepancy

partly arises from the different temporal allocation of emissions in the two inventories. Temporally, THRIVE-SO₂ releases SO₂ progressively into the atmosphere. In contrast, VolcanEESM prescribes the total eruptive SO₂ mass at a constant rate over a fixed 6-h period, producing a more abrupt increase in simulated SO₂ mass and limiting its representation of eruptions with more prolonged or temporally variable SO₂ emissions. The different vertical allocation of emissions in the two inventories compounds this discrepancy. THRIVE-SO₂ resolves the altitude-dependent structure of the emissions, whereas VolcanEESM distributes SO₂ uniformly by volume between the reported lower and upper plume altitudes. As a result, the simulated plumes in the two inventories encounter different meteorological and chemical conditions, contributing to the differences in the subsequent evolution and depletion of the SO₂ burden.

In particular, for the Merapi case, VolcanEESM does not provide SO₂ emission information until 8 November 2010, after the eruption had ended (Fig. 2b). This delayed, temporally aggregated emission input further amplifies differences in the evolution of the SO₂ burden and its subsequent conversion to sulfate aerosol.

The upgraded PCA algorithm²⁶ used to derive the OMI column amount of volcanic SO₂ still underestimates SO₂ loading for very dense plumes immediately following large eruptions, as observed during the Nabro eruption. Furthermore, OMI provided little to no valid coverage on 15 June 2011, precluding a meaningful comparison for this date. In addition, for eruptions that emit large amounts of volcanic ash, such as the Kelud eruption, the ash may interfere with SO₂ retrieval, leading to an underestimation of the retrieved SO₂ mass.

To more clearly show how different emission inventories affect simulated plume structure and transport pathways, we use the Nabro eruption as an example and examine the corresponding SO₂ plume evolution in space and time. Figure 3 compares the spatial evolution of the SO₂ plume from the Nabro eruption as reproduced by two model simulations initialized with THRIVE-SO₂ and VolcanEESM, against AIRS satellite observations.

On 13 June 2011, shortly after the eruption onset, both THRIVE-SO₂- and VolcanEESM-initialized simulations reproduce a compact SO₂ plume centered near the Nabro volcano (marked by the red triangle) that closely matches the confined signal seen in the AIRS retrieval. As the plume is advected downstream and progressively disperses over the following days, the differences between these two simulations become apparent. Generally, the VolcanEESM simulation keeps the SO₂ plume much more spatially concentrated, whereas the THRIVE-SO₂ produces substantially stronger dispersion. For the VolcanEESM-initialized simulation, the plume trajectory and spatial spreading quickly depart from the AIRS observations. In contrast, the THRIVE-SO₂ simulation continues to reproduce SO₂ plumes much more consistent with the spatial extent and structure seen in the AIRS observations. It correctly captures the plume's transport pathway, horizontal extent, and gradual depletion, with only minor discrepancies in

fine-scale structure likely attributable to sub-grid transport processes and differences in observational sampling times.

This suggests that the deviations arising from the simplified representation of the initial SO₂ emission in VolcanEESM are amplified as transport proceeds. Because the total SO₂ mass allocated is identical between the THRIVE-SO₂ and VolcanEESM-initialized simulations, the more spatially compact plume in the VolcanEESM simulation consistently exhibits a higher SO₂ column density than the THRIVE-SO₂ simulation.

Overall, the comparison indicates that the THRIVE-SO₂-initialized simulation exhibits substantially better agreement with the observed distribution of the SO₂ plume than the VolcanEESM-initialized simulation, particularly in reproducing the plume's transport pathway and dispersion pattern at later stages of the SO₂ plume evolution. It should be noted that the THRIVE-SO₂ is derived from AIRS observations in combination with inverse modeling techniques, and this inherent data dependency may partly explain the closer agreement with the AIRS-observed plume distribution.

A similar pattern of better spatial agreement with AIRS observations for the THRIVE-SO₂ simulation is also found for the other four eruption cases examined in this study. The corresponding plume evolution comparisons are provided in Supplementary Figs. 2–5.

To quantitatively evaluate the spatial agreement between the simulated and observed SO₂ plumes, we calculated the critical success index (CSI)^{27–28}. First, we gridded the model output at 06:00 UTC each day onto a 1°×1° latitude–longitude grid, along with the corresponding AIRS SO₂ retrievals within ± 6 h of that time. Each grid cell in the simulation and satellite observation domain was then classified as a positive or negative detection based on a prescribed SO₂ threshold of 3 DU in this study. The outcomes are tallied into three categories: correctly simulated plume presence (C_x), plume presence missed by the model (C_y), and plume falsely simulated by the model where none was observed (C_z).

The probability of detection (POD):

$$POD = \frac{C_x}{C_x + C_y}$$

1

,

quantifies the fraction of the observed plume area that is successfully reproduced by the simulation.

The false alarm rate (FAR):

$$FAR = \frac{C_z}{C_x + C_z}$$

2

,

quantifies the fraction of the modeled plume area that has no corresponding observed signal.

The miss rate (MIS):

$$MIS = 1 - POD$$

3

,

reflects the complementary fraction of missed plume area.

The CSI combines these two aspects of model skill into a single score:

$$CSI = \frac{C_x}{C_x + C_y + C_z}$$

4

,

which ranges from 0 (no spatial overlap between simulated and observed plumes) to 1 (perfect spatial agreement), providing an overall measure of how well the modeled plume footprint matches the satellite-observed distribution at each verification time.

(a)

Figure 4. Time series of the performance metrics of probability of detection (POD), false alarm rate (FAR), miss rate (MIS), and critical success index (CSI) for Sarychev, Merapi, and Nabro eruptions. The column density threshold used to detect events is 3 DU. The red and yellow solid lines show the performance metrics of SO₂ transport simulations initialized with THRIVE-SO₂ and VolcanEESM, with AIRS SO₂ retrievals as the reference. The gray lines show the performance metrics of the SO₂ transport simulation initialized with VolcanEESM, with the THRIVE-SO₂-initialized simulation used as the reference.

(a)

Figure 5. Same as Fig. 4, but for the Kelud and Calbuco eruptions.

Figure 4 and Fig. 5 present the time series of POD, FAR, MIS, and CSI computed for the SO₂ plume simulations of all five eruption cases. The red and yellow lines denote the performance of THRIVE-SO₂- and VolcanEESM-initialized simulations relative to AIRS, respectively, while the gray lines show the performance of the VolcanEESM simulation using the THRIVE-SO₂ simulation as the reference. Consistent with the spatial comparisons in Fig. 3, the THRIVE-SO₂-initialized simulation exhibits higher

POD and CSI values, together with lower MIS values, than the VolcanEESM-initialized simulation across nearly all cases, indicating that the plume of the THRIVE-SO₂ simulation overlaps more closely with the AIRS-observed plume.

The POD and CSI are largest during the first few days after the eruption. Then, as the plume becomes more dilute and fragmented, POD decreases but MIS and FAR increase at later stages. The increase in MIS and FAR reflects the accumulation of small differences in transport pathways and dispersion as the plume moves downstream. These differences reduce overlap between the simulated and observed plumes.

The gray lines directly quantify the discrepancies between simulations initialized with VolcanEESM and those initialized with THRIVE-SO₂. As shown by the Sarychev and Nabro cases, relatively low POD and FAR values combined with high MIS indicate that the VolcanEESM plume is narrower than the THRIVE-SO₂ plume and therefore misses a larger fraction of it. In contrast, during the later stages of the Kelud and Calbuco cases, high POD and FAR values together with low MIS indicate that the VolcanEESM plume encompasses part of the THRIVE-SO₂ plume but extends beyond it, reflecting differences in plume transport pathways and horizontal dispersion between the two simulations.

As mentioned earlier, VolcanEESM does not provide SO₂ emissions for the Merapi case until after the eruption ended. Consequently, the CSI of the VolcanEESM simulation for the Merapi case remained zero throughout the entire simulation period.

Comparison of volcanic aerosol transport

Since the climatic impact of volcanic eruptions is ultimately governed by the sulfate aerosol formed after the eruptions, we next evaluate the simulated stratospheric sulfate aerosol loading. We used the MPTRAC model, driven separately by THRIVE-SO₂ and VolcanEESM, to simulate the transport and dispersion of sulfate aerosols over the six months following each eruption, and compared the results against the 525 nm CALIOP aerosol extinction coefficients from the Global Space-based Stratospheric Aerosol Climatology (GloSSAC)²⁹. As noted in the last section, except for the different temporal and spatial distribution of the initial SO₂ emission, both simulations used the same model configuration, including the same number of particles (100,000) and the same total SO₂ mass, with each particle carrying an equal initial sulfur mass.

Figure 6 presents the latitude–altitude evolution of aerosol loading following the Nabro eruption, comparing the 525 nm CALIOP aerosol extinction coefficients from GloSSAC (left column) with the simulated aerosol particle number from the THRIVE-SO₂ (middle column) and the VolcanEESM (right column) for July, September, and November 2011. The CALIOP extinction coefficients represent all stratospheric aerosols detectable by the instrument, including background aerosol, whereas the model

simulations represent only the sulfate aerosol originating from the volcanic eruption; this distinction should be kept in mind when interpreting the absolute magnitude of the comparison.

As shown in Fig. 6, the CALIOP observations show a pronounced enhancement of aerosol loading in the Northern Hemisphere lower stratosphere shortly after the eruption, which is explained by the volcanic plume being transported first to the Northern Hemisphere along the anticyclonic circulation of the Asian summer monsoon⁷, as shown in Fig. 3. From July to November 2011, the observed aerosol loading gradually decreases in magnitude, while a fraction of the aerosol is progressively transported quasi-horizontally toward the tropical UTLS region. Some upward-spreading traces are also discernible within the tropical lower stratosphere, consistent with the tropical upwelling.

The THRIVE-SO₂-initialized simulation reproduces this observed evolution well, capturing both the initial mid-latitude aerosol enhancement and its subsequent equatorward transport and gradual decay, in good qualitative agreement with the CALIOP observations. The VolcanEESM-initialized simulation reproduces a broadly similar transport pattern, but with a distinctly larger tropical lower-stratospheric aerosol abundance and a stronger subsequent upward extension of the aerosol layer. This discrepancy likely arises because the VolcanEESM injects a larger fraction of SO₂ at higher altitudes. As a result, the VolcanEESM-driven simulation may overestimate both the amount of volcanic aerosol entrained into the Brewer-Dobson circulation and its residence time in the stratosphere, which may in turn bias the assessment of the Nabro eruption's climatic impact.

Figure 7 presents the same latitude–altitude comparison as Fig. 6, but for the Kelud eruption in 2014. The CALIOP observations reveal that the Kelud volcanic aerosol, initially appearing near the tropical UTLS, is subsequently transported quasi-horizontally toward the Southern Hemisphere lower stratosphere over the following months, forming an arch-shaped aerosol layer that extends from the tropics into southern mid-latitudes by May and July 2014.

The THRIVE-SO₂-driven simulation captures this quasi-horizontal transport pattern reasonably well, reproducing both the tropical aerosol enhancement shortly after the eruption and its gradual poleward extension into the Southern Hemisphere, broadly consistent with the CALIOP observations. In contrast, the VolcanEESM-driven simulation exhibits a markedly different vertical distribution: because the VolcanEESM injects all of the SO₂ mass essentially within a narrow altitude range of 17–26 km, the resulting stratospheric sulfate aerosol loading is substantially overestimated near the tropical UTLS. This excess aerosol mass simultaneously enhances both quasi-horizontal transport toward the lower stratosphere of the Southern Hemisphere and vertical uptake into the ascending branch of the Brewer-Dobson circulation. This suggests that the VolcanEESM-based simulation likely overestimates the stratospheric residence time and hemispheric spread of Kelud volcanic aerosol, which could introduce corresponding biases in, for example, estimates of stratospheric radiative forcing and in assessments of Antarctic stratospheric chemistry.

Similar comparisons for the remaining three eruption cases, the Sarychev, Merapi, and Calbuco eruptions, are provided in Supplementary Figs. 6–8. Across these cases, the THRIVE-SO₂-initialized simulations consistently reproduce the observed CALIOP aerosol distribution more closely than the VolcanEESM-initialized simulations, although the degree of improvement varies with the specific injection characteristics and transport pathway of each eruption.

Discussion

This study developed a high-resolution, altitude- and time-resolved volcanic SO₂ emission inventory, THRIVE-SO₂, covering 52 eruptions from 2002 to the present, and assessed its performance against a conventional, coarsely resolved inventory (the VolcanEESM) across five selected eruptions spanning a range of latitudes, injection altitudes, and dynamical regimes.

These results demonstrate that THRIVE-SO₂ substantially improves the simulated accuracy of SO₂ transport, depletion, and sulfate aerosol dispersal. These improvements stem from two related capabilities that conventional inventories lack. First, by resolving the accurate temporal evolution of SO₂ release, the THRIVE-SO₂ captures the gradual, often multi-day buildup of a volcanic plume, rather than approximating the eruption as an instantaneous or uniformly distributed release over a fixed short window. This distinction proved especially consequential for eruptions with a protracted SO₂ release, where a rigid emission window can cause simulations to miss the plume's early evolution. Second, rather than relying on a roughly estimated altitude range uniformly filled with SO₂, as in the VolcanEESM, the THRIVE-SO₂ reconstructs the vertical distribution of SO₂ directly from satellite-based trajectory retrievals. This approach allows THRIVE-SO₂ to separate SO₂ more realistically between the troposphere, the UTLS region, and the stratosphere. Because these three regimes differ fundamentally in their wind fields, chemical environments, and removal timescales, even modest errors in this vertical partitioning can propagate into substantial differences in simulated aerosol lifetime and spatial spread.

It should be acknowledged that the VolcanEESM captures most of the SO₂ mass in the upper troposphere and stratosphere, the altitude range most relevant to climate impact. As a result, it remains broadly sufficient for many climate modeling applications. THRIVE-SO₂, however, provides a substantially more detailed and realistic representation of the volcanic SO₂ emission process and improves SO₂ transport, depletion, and sulfate aerosol dispersal simulation.

In summary, this study shows that the improved vertical and temporal resolution of the volcanic SO₂ emission inventory substantially improved consistency between simulated SO₂ plumes and satellite observations, underscoring the value of high-resolution emission inventories for eruption impact assessment. Future work will focus on the continued refinement and updating of the THRIVE-SO₂, as well as its implementation in the WACCM model to investigate the broader climate impacts of volcanic eruptions. In particular, this framework will be used to examine the response of polar stratospheric

ozone to volcanic forcing, providing a more comprehensive assessment of the utility of THRIVE-SO₂ in fully coupled climate simulations.

Method

Lagrangian particle dispersion model: MPTRAC

The transport, dispersion, and chemical evolution of volcanic SO₂ in this study are simulated using the Massive-Parallel Trajectory Calculations model (MPTRAC) version 2.6²⁵. MPTRAC is a Lagrangian particle dispersion model originally developed to study transport processes in the free troposphere and stratosphere³⁰, with particular applications to tracking the long-range transport of volcanic SO₂ and sulfate aerosols. The MPTRAC model has been widely applied to investigate the long-range transport and dispersion of volcanic SO₂ and sulfate aerosols from a range of eruption events^{6–7, 13, 16, 18}.

MPTRAC calculates the trajectories of a large ensemble of individual particles by solving the kinematic equation of motion, driven by horizontal wind and vertical velocity fields taken from global reanalysis or forecast products. In this study, the model is driven by hourly ERA5 reanalysis data³¹. To account for subgrid-scale turbulence and unresolved wind fluctuations, MPTRAC applies a stochastic diffusion scheme based on the Langevin equation, supplemented by an inter-particle exchange module that represents small-scale air mixing between neighboring trajectories. Unresolved convective transport is represented through an extreme convection parameterization³⁰.

Beyond physical transport, MPTRAC incorporates a suite of chemical and microphysical modules relevant to volcanic SO₂ plume evolution, including gravitational sedimentation, gas-phase and aqueous-phase chemistry, and both wet and dry deposition. The chemical decay of SO₂ is represented through gas-phase oxidation by the hydroxyl radical (OH) and, within cloud regions, through aqueous-phase oxidation by hydrogen peroxide (H₂O₂) as well as in-cloud wet scavenging, with reaction rates and scavenging coefficients formulated as functions of temperature, pressure, cloud water content, and precipitation rate derived from the meteorological input data. These improvements in Version 2.6 substantially improve the realism of simulated SO₂ depletion and the corresponding formation of sulfate aerosol compared to earlier model versions¹⁸.

MPTRAC is optimized for high-performance computing environments through a hybrid MPI-OpenMP-OpenACC parallelization scheme, enabling efficient large-scale simulations with up to hundreds of millions of air parcels or particles on both CPU and GPU architectures³⁰.

For each volcanic eruption case, 100,000 particles were initialized to represent the emitted SO₂ for each volcanic eruption case. To enable a direct and consistent comparison, the initial total SO₂ mass of the THRIVE-SO₂ was scaled to match that of the VolcanEESM for the five eruptions examined in this study. Each particle was assigned an equal initial SO₂ mass. Trajectories initialized with the SO₂ emission time

series from either THRIVE-SO₂ or VolcanEESM were then simulated for six months using the MPTRAC model.

Satellite measurements

To evaluate the simulated transport of the volcanic SO₂ plume using the Lagrangian particle dispersion model, we use SO₂ retrievals from AIRS, an infrared sounder aboard the Aqua satellite launched in May 2002. AIRS completes roughly 14.5 orbits per day, achieving near-global coverage twice daily, with a nadir footprint of approximately 13.5 × 13.5 km.

Volcanic SO₂ is identified through a brightness temperature (BT) difference approach in the 7.3 μm absorption band. Specifically, the SO₂ index (SI) is calculated as the difference between BT at 1412.87 cm⁻¹ and a reference channel at 1371.52 cm⁻¹:

$$SI = BT(1412.87 \text{ cm}^{-1}) - BT(1371.52 \text{ cm}^{-1}). \quad (5)$$

The SO₂ detection approach follows Hoffmann et al. (2014)³², using an optimized selection of AIRS channels that improves sensitivity to weak volcanic SO₂ signals and reduces background interference compared with the operational NASA AIRS SO₂ index. The resulting AIRS SI is most sensitive to SO₂ in the UTLS region, approximately 8–13 km altitude. The SO₂ index from AIRS detections was then converted into SO₂ column density using the correlation function described in Hoffmann et al. (2014)³², which was obtained from radiative transfer calculations.

In this study, the AIRS SO₂ observations are used to estimate the volcanic SO₂ emission time series together with the MPTRAC model, and are used to compare with the model-simulated SO₂ column density.

In addition, OMI SO₂ measurements are used to evaluate the volcanic SO₂ mass simulated by the MPTRAC model. OMI is on board the Aura satellite, launched in July 2004. OMI is an ultraviolet/visible (UV/VIS) nadir solar back-scatter spectrometer with daily global coverage. The instrument has a spatial resolution of 13 km × 24 km at nadir. The SO₂ retrievals are derived from the 310.5–340 nm range of OMI's UV2 channel, with particular sensitivity to the 310.8 and 313 nm wavelengths²⁶. In this study, we use the OMSO2 v004 product³³ and convert the retrieved SO₂ column concentrations to SO₂ mass for direct comparison with the simulation results.

In addition, the Global Space-based Stratospheric Aerosol Climatology (GloSSAC)²⁹ is used to evaluate the temporal evolution of stratospheric aerosols simulated by the MPTRAC model. GloSSAC is a long-term, multi-satellite compilation of stratospheric aerosol properties developed and maintained by NASA, merging observations from a series of satellite-borne instruments spanning from 1979 to the present. The dataset provides monthly zonal-mean stratospheric aerosol extinction coefficients at multiple

wavelengths, with a spatial resolution of 5° in latitude and 0.5 km in altitude, extending from the tropopause to approximately 40 km. In this study, we specifically use the 525 nm aerosol extinction coefficients derived from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) observations incorporated in GloSSAC, given their comprehensive temporal and spatial coverage over the study period. These CALIOP-derived extinction coefficients serve as observational references for assessing the vertical and latitudinal distributions of volcanic aerosols simulated by the MPTRAC model.

Declarations

CODE AVAILABILITY

The code for the Massive-Parallel Trajectory Calculations (MPTRAC) model is available under the terms and conditions of the GNU General Public License, Version 3, from <https://github.com/slcs-jsc/mptrac> (last accessed: 21 April 2026).

COMPETING INTERESTS

The authors declare no competing interests.

Author Contribution

X. W., L. H., and B. C. developed the idea. All the authors discussed the concepts. X. W. and W. Z. performed most of the analysis. X. W. drafted the manuscript, and all authors edited the manuscript.

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Data Availability

The VolcanEESM global volcanic SO₂ emissions database is available at <https://data.ceda.ac.uk/badc/volcan-eesm/data/v1> (last access: 18 August 2026). The high-resolution, altitude-resolved volcanic SO₂ emission inventory (the THRIVE-SO₂) is available at https://datapub.fz-juelich.de/slcs/so2_inv/index.html (last access: 21 April 2026). The AIRS SO₂ index data are available for download at <https://datapub.fz-juelich.de/slcs/airs/volcanoes/> (last access: 21 April 2026). The SO₂ measurements from OMI are available at https://disc.gsfc.nasa.gov/datasets/OMS02_004/ (last access: 21 April 2026). The Global Space-based Stratospheric Aerosol Climatology (GloSSAC) data is available at <https://www.earthdata.nasa.gov/data/projects/glossac> (last accessed: 02 August 2016). The data used to generate the figures are available at <https://doi.org/10.5281/zenodo.22854293> (last access: 20 September 2026).

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Figures

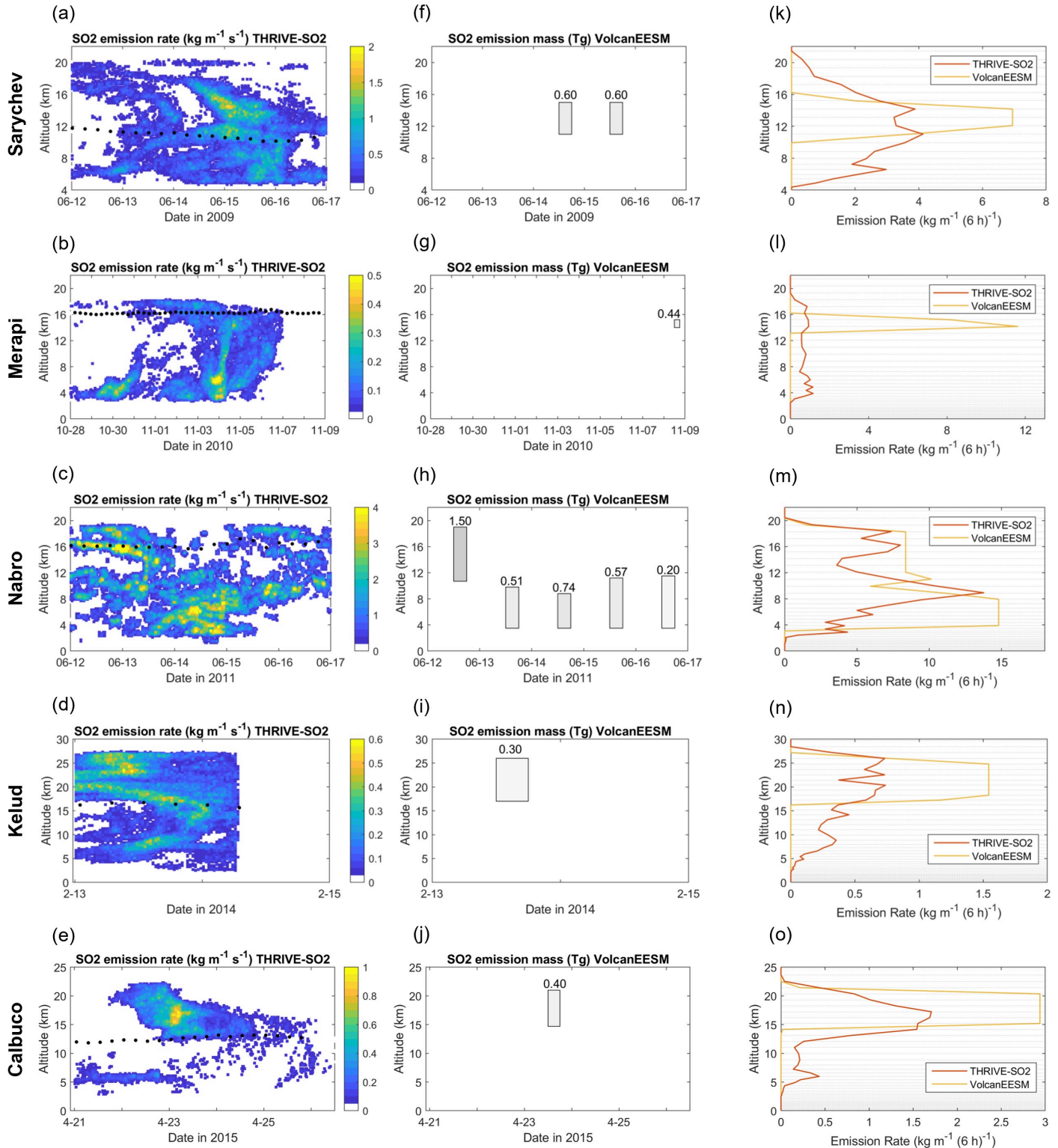


Figure 1

SO₂ emission time series and vertical profiles of the Sarychev eruption (2009), Merapi eruption (2010), the Nabro eruption (2011), the Kelud eruption (2014), and the Calbuco eruption (2015). (a)–(e) are SO₂ emissions from THRIVE-SO₂. The bin size is 15 minutes × 0.2 km. Black dots indicate tropopause height. (f)–(j) are SO₂ emissions from VolcanEESM. Panels (k)–(o) compare the vertical profiles of the SO₂

emission integrated over the eruption period and interpolated onto the model levels of the WACCM model, indicated by the light gray horizontal lines.

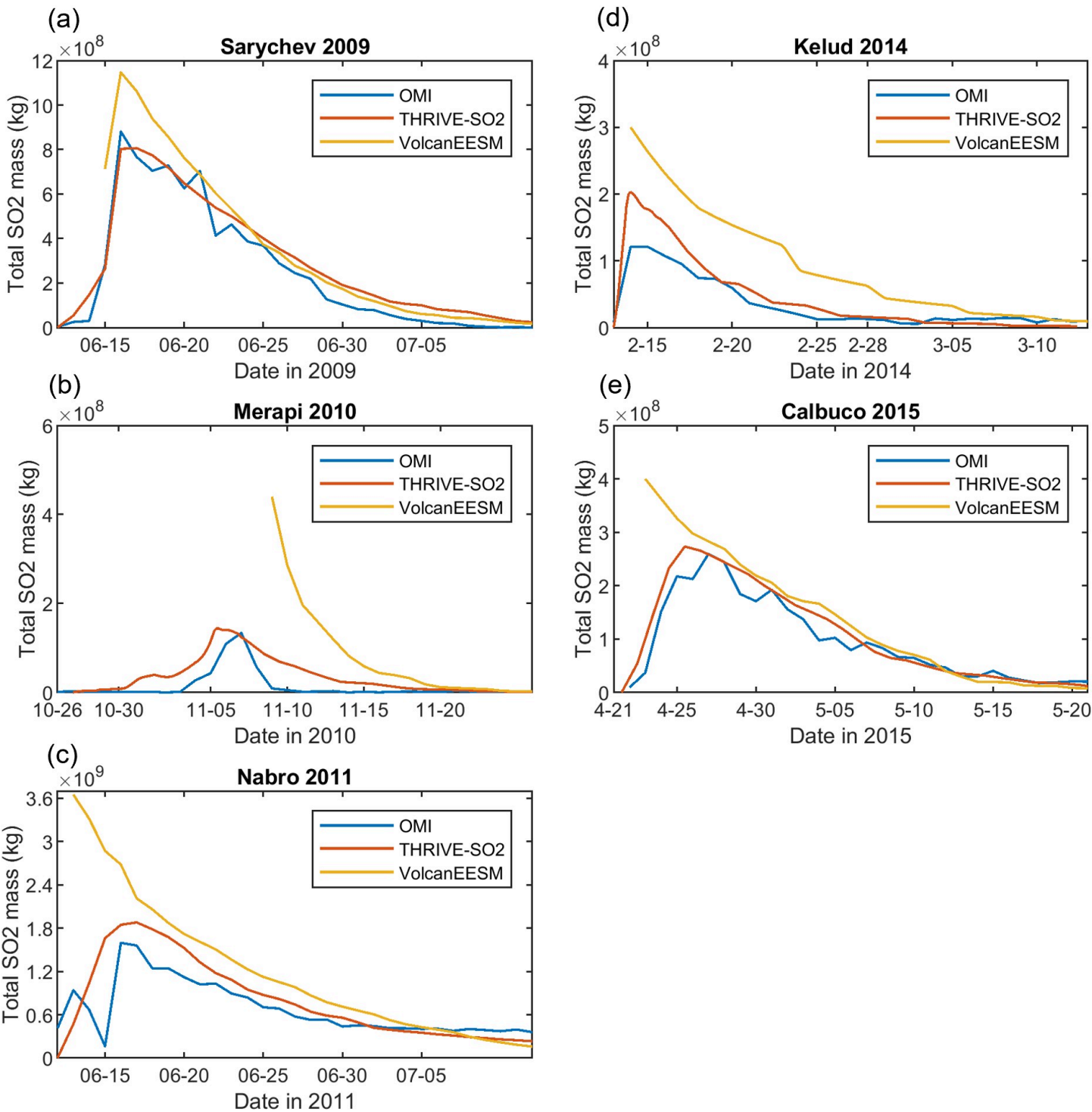


Figure 2

Comparison of the simulated total SO₂ mass time series initialized by the THRIVE-SO₂ and VolcanEESM against OMI SO₂ observations for each eruption case.

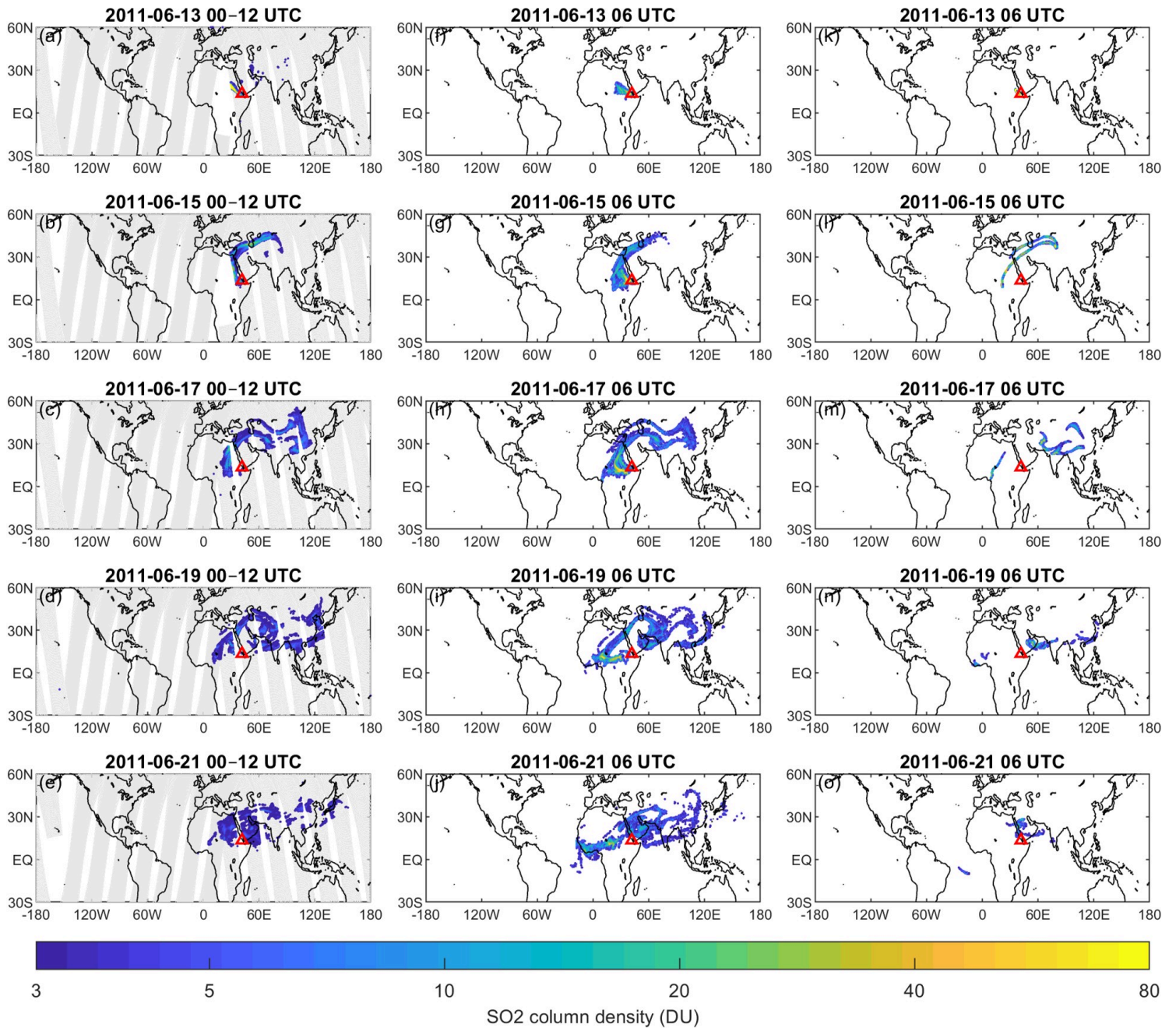


Figure 3

Comparison of the simulated SO_2 column density evolution initialized by the THRIVE- SO_2 and the VolcanEESM against AIRS SO_2 column density observations for the Nabro eruption from 13 to 21 June 2011. The MPTRAC SO_2 column densities are shown at 06:00 UTC on selected days, and AIRS data within ± 6 h are shown. The red triangle denotes the location of the Nabro volcano.

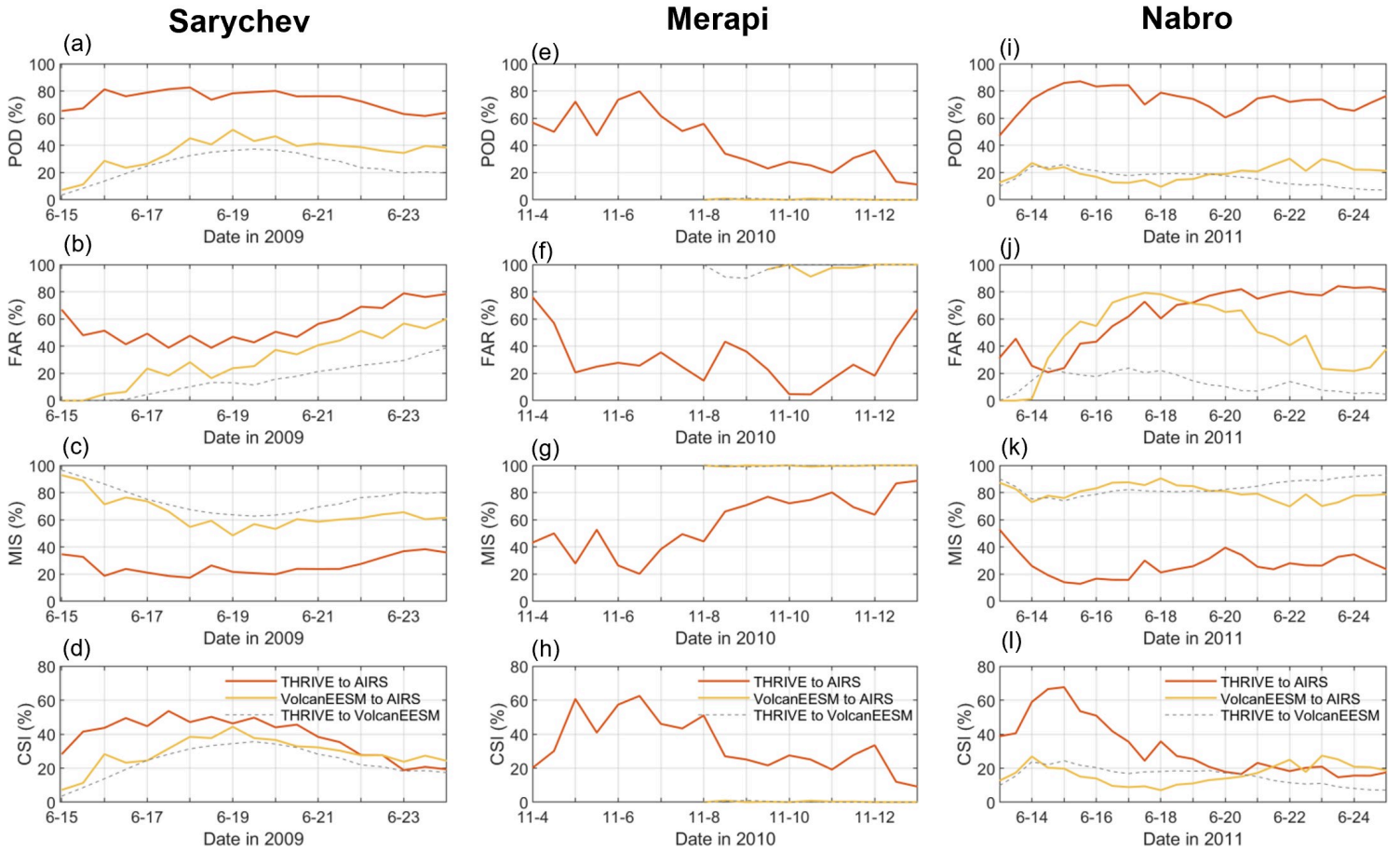


Figure 4

Time series of the performance metrics of probability of detection (POD), false alarm rate (FAR), miss rate (MIS), and critical success index (CSI) for Sarychev, Merapi, and Nabro eruptions. The column density threshold used to detect events is 3 DU. The red and yellow solid lines show the performance metrics of SO_2 transport simulations initialized with THRIVE- SO_2 and VolcanEESM, with AIRS SO_2 retrievals as the reference. The gray lines show the performance metrics of the SO_2 transport simulation initialized with VolcanEESM, with the THRIVE- SO_2 -initialized simulation used as the reference.

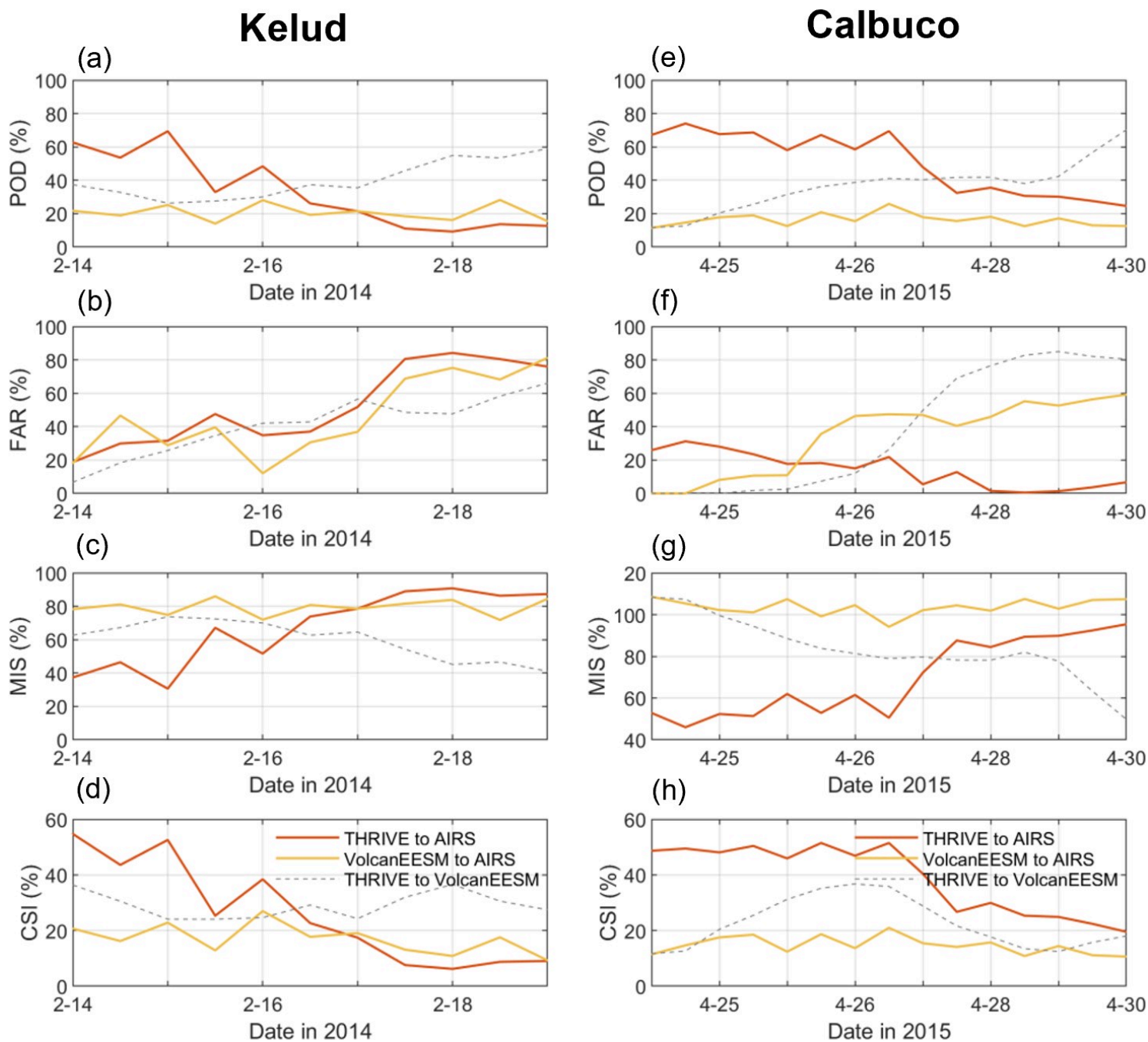


Figure 5

Same as Fig. 4, but for the Kelud and Calbuco eruptions.

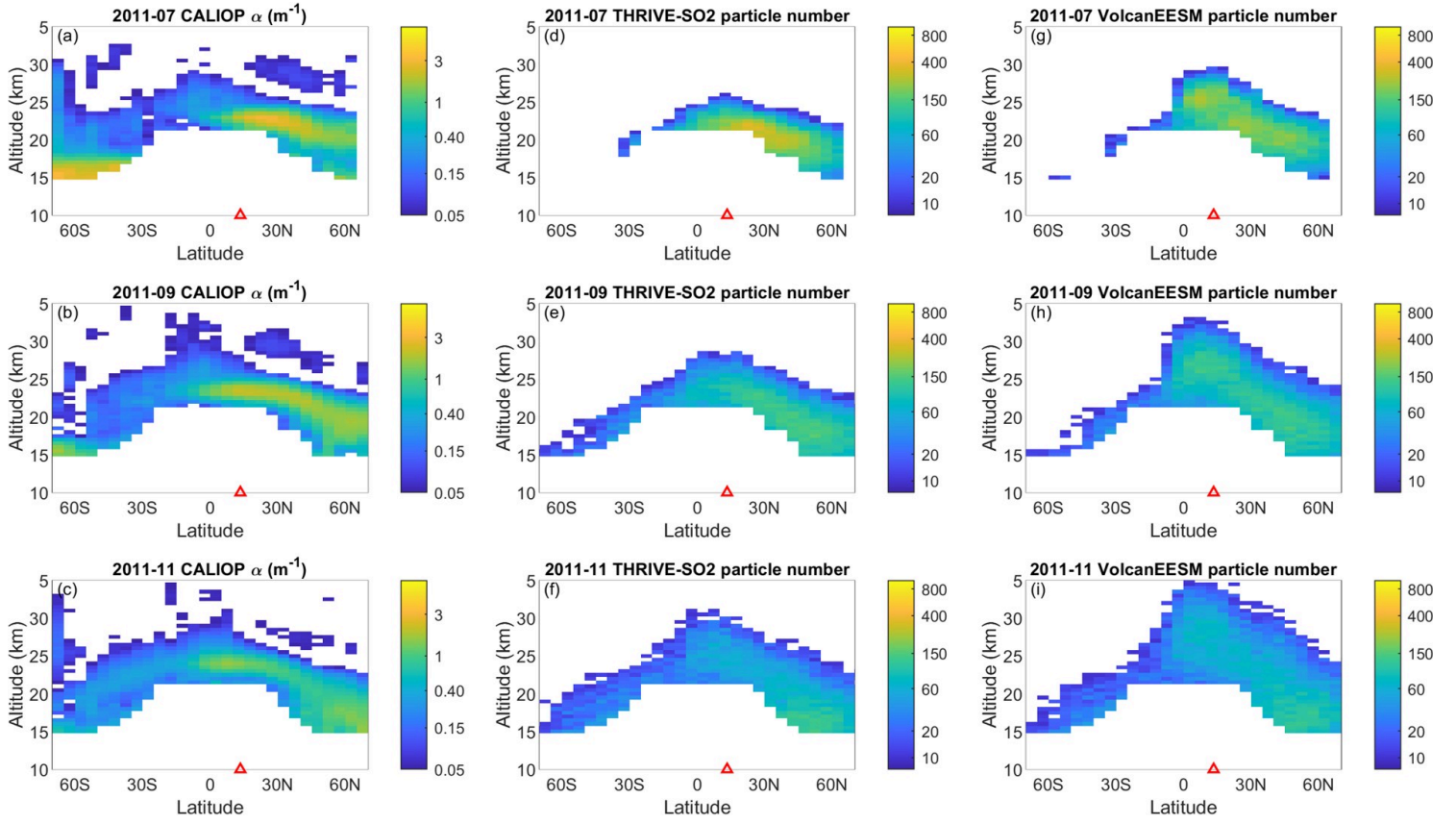


Figure 6

Latitude-altitude cross-sections of aerosols from July 2011 to November 2011 following the Nabro volcano eruption in June 2011. Left column: the 525 nm CALIOP aerosol extinction coefficients from the Global Space-based Stratospheric Aerosol Climatology (GloSSAC); Middle column: simulated aerosol particle count initialized by the THRIVE-SO₂; Right column: simulated aerosol particle count initialized by the VolcanEESM. The bin size is 5°×0.5 km. The red triangles indicate the latitude of the Nabro volcano. Regions with missing CALIOP data are masked in the simulations for better comparison.

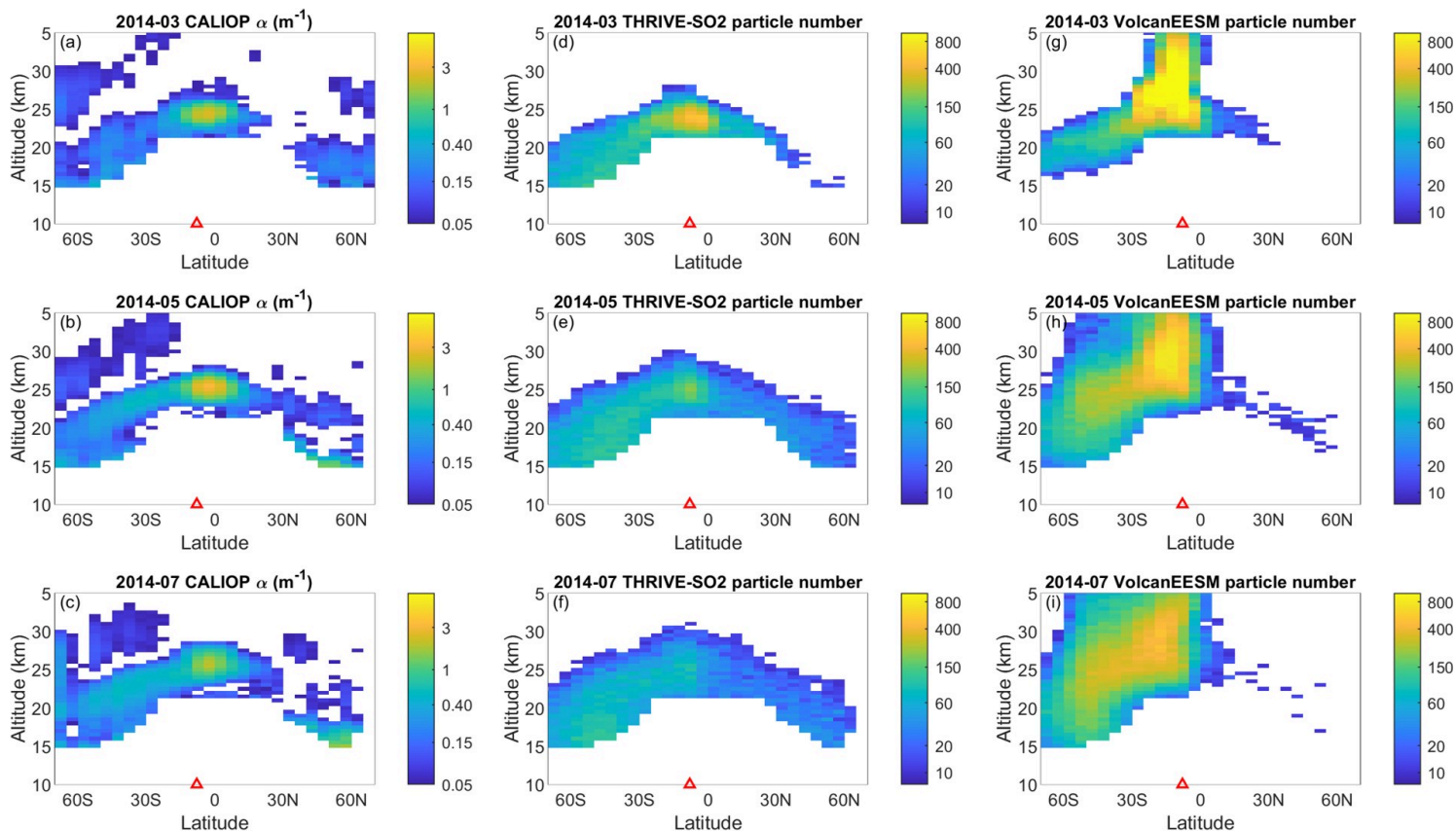


Figure 7

Same as Fig. 6 but for the Kelud eruption in 2014.

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