~~Radiative forcing by~~ Reconstructing volcanic ~~eruptions~~ radiative forcing since 1990, ~~calculated with~~ using a ~~chemistry-climate model and a new~~ comprehensive emission inventory ~~based on vertically and spatially~~ resolved sulfur injections from satellite ~~measurements~~ data in a chemistry-climate model

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Abstract.

This paper presents model simulations of stratospheric aerosols with a focus on explosive volcanic eruptions. Using various (~~occultation and limb based~~occultation and limb-based) satellite instruments, with vertical profiles of sulfur dioxide (SO_2) ~~from the MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) instrument~~ and vertical profiles of aerosol extinction~~from GOMOS (Global Ozone Monitoring by Occultation of Stars), OSIRIS (Optical Spectrograph and InfraRed Imaging System), and SAGE II (Stratospheric Aerosol and Gas Experiment), we characterised~~, we characterized the influence of volcanic aerosols for the period between 1990 and 2019.

We established ~~a~~ an improved and extended volcanic sulfur emission inventory that includes more than 500 ~~eruptions .~~ ~~The identified~~ explosive volcanic eruptions reaching the upper troposphere and the stratosphere. Each perturbation identified was derived directly from the satellite data and incorporated as a three-dimensional SO_2 ~~perturbations were incorporated as three-dimensional pollution plumes~~ plume into a chemistry-climate model~~, which converts the gases~~. The simultaneous measurements of SO_2 and aerosol extinction by up to four instruments enabled us to develop a reliable method to convert extinction measurements into injected SO_2. In the chemistry climate model, the SO_2 from each individual plume is converted into aerosol particles and ~~computes~~ their optical properties ~~. The~~ are determined. Furthermore, the Aerosol Optical Depth (AOD) and the ~~climate radiative forcing~~ instantaneous radiative forcing on climate are calculated online. Combined with model improvements, the simulations ~~reproduce~~ are consistent with the observations of the various satellites.

Slight deviations between the observations and model simulations were found ~~only~~ for the large volcanic eruption of Pinatubo in ~~1991. This is likely due to either an overestimation of the removal of aerosol particles in the model, or limitations of the satellite measurements, which are related to saturation effects associated with anomalously high aerosol concentrations. Since Pinatubo, only smaller-sized volcanic eruptions have taken place.~~ 1991 and cases where simultaneous satellite observations

were not unique or too sparse. Weak- and medium-strength volcanic eruptions captured in satellite data and the Smithsonian database typically inject about 10 kt to 50 kt SO_2 directly into the upper troposphere/lower stratosphere (UTLS) region or transport it indirectly via convection and advection. Our results ~~show~~ confirm that these relatively smaller eruptions, which occur quite frequently, can nevertheless contribute ~~significantly~~ to the stratospheric aerosol layer and are relevant for the Earth's radiation budget. ~~These eruptions are found to~~ These eruptions cause a global radiative forcing ~~in~~ of the order of $-0.1\,\mathrm{Wm^{-2}}$ at the tropopause ~~.~~ (compared to a background aerosol forcing of about $-0.04\,\mathrm{Wm^{-2}}$).

1 Introduction

~~Excluding recent history~~

Next to recent historical events in which large fires have become a major source of aerosols up to the tropopause and above it (Kloss et al., 2019), stratospheric aerosol particles are mostly of volcanic origin and consist of a mixture of liquid sulfuric acid (H_2SO_4) and water (H_2O) droplets ~~, which are condensed from the gas phase by secondary particle formation.~~ (Vernier et al., 2011). The typical median diameter of these aerosol particles ranges between 200 nm for the background aerosol and 600 nm for Pinatubo conditions ~~(Brühl et al., 2015). As species with a relatively long lifetime, stratospheric aerosol particles show a tendency to accumulate in the lower stratospheric aerosol layer at altitudes of between and (?). At these heights, they scatter the incoming solar radiation in the ultraviolet (UV), visible and near-infrared (IR) ranges and absorb the outgoing terrestrial mid-infrared radiation, in this way influencing the radiation budget of the Earth's atmosphere.~~ (Wilson et al., 2008). In this study, we incorporated stratospheric aerosol, ~~including the sulfur budget, the sulfur~~ the sulfur chemistry and the radiative transfer into a comprehensive Chemistry Climate Model (CCM), which we have used to gain a better understanding of the interaction of aerosols with the global climate system~~and ozone.~~, including chemical effects. Much focus is ~~currently~~ being placed on adequately modelling and understanding the impact of volcanic eruptions and other aerosol sources on the evolution of the stratospheric aerosol burden.

Sulfate and ashes from explosive volcanic eruptions can account for the majority of the aerosol burden in the stratosphere during volcanically active periods and cause strong temporal and spatial variations in the concentration and the size distribution of the particles ~~.~~ (Vernier et al., 2011). These changes influence in turn the radiative forcing at tropopause altitudes (or at the top of the atmosphere) for several years after strong eruptions (Timmreck, 2012) and can even have a ~~large~~ more prolonged impact on the global climate ~~for one to three years after the eruptions (Timmreck, 2012).~~ (McGregor et al., 2015). After such a volcanic eruption, the enhanced radiative heating exerts an effect on dynamics, influences the global spread of the volcanic cloud and leads to an upward transport of the aerosol itself as well as of other chemical tracers including ozone (Timmreck et al., 1999). The aerosol radiative heating resulting from large volcanic eruptions like Pinatubo triggers enhanced tropical upwelling, which causes a lofting of the injected SO_2 (sulfur dioxide) and the aerosol as well as other chemical tracers. The radiative feedback on dynamics is required to model aerosol extinction in the upper part of the volcanic aerosol plume that corresponds with observations.

Due to the large variability in volcanic emissions, it is ~~not possible to define~~ challenging to estimate future trends for strato-spheric optical depth and forcing ~~.~~ (Swindles et al., 2018; Fasullo et al., 2017; Aubry et al., 2021). Therefore, the influence of volcanic eruptions is not included in predictive simulations for future climate scenarios in the IPCC report from 2013 (IPCC, 2013). Previous studies show that model simulations often cannot completely reproduce the AOD of satellite observations or the global forcing of the stratospheric aerosol layer (Solomon et al., 2011), because the number of volcanic eruptions reaching the stratosphere is underestimated in most analyses ~~(Mills et al., 2016; Brühl et al., 2015).~~ (Mills et al., 2016; Brühl et al., 2015; Schmidt et al., . Conversely, the intensity of single eruptions is sometimes overestimated because of incorrect vertical distribution of the injec-tion patterns in the models. Smaller volcanic eruptions have often been underestimated in previous studies~~(Brühl et al., 2015)~~ ~~.,~~ even though they can be responsible for a radiative forcing that is twice as strong as the background conditions in volcanically quiescent periods such as from 1999 to 2002 ~~(IPCC, 2013). Therefore, sulfur dioxide (~~ (IPCC, 2013; Solomon et al., 2011; Vernier et al., 20 . Friberg et al. (2018) included the whole time series of CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) data from 2006 to 2015 and derived stratospheric AOD using reanalysis data for the tropopause, but mentions only medium size eruptions explicitly. Radiative forcing is estimated there from multiplying AOD with -25, an approach which is valid only for purely scattering aerosol. GLOSSAC (Thomason et al., 2018; Kovilakam et al., 2020), a time dependent aerosol climatology sometimes used for climate simulations, has a coarse temporal resolution, many discontinuities in space and time due to change of instruments or measurement gaps and excludes important satellite data (e.g. MIPAS). It does not provide SO_2 ~~)~~needed as input for chemistry climate models directly but only extinction and indirectly derived quantities like estimates of aerosol surface area density and mode radius assuming unrealistic monomodal size distributions. We therefore prefer to recalculate SO_2 injections into the stratosphere ~~from previously underestimated and unknown volcanic eruptions have to be recalculated~~ based on the Smithsonian volcano database and the most recent releases of satellite data sets, in particularly those gathered using limb sounding instruments to derive information ~~in the vertical direction.~~ on the vertical distribution.

For the ENVISAT (European Environmental Satellite) period 2002-2012 a first version of a new volcanic SO_2 inventory with improved temporal and spatial resolution was developed within the framework of ISA-MIP (Timmreck et al., 2018; Brühl et al., 2018) . The corresponding data base (https://doi.org/10.1594/WDCC/SSIRC_1) contains 3D-SO_2-perturbations derived from satellite data as well as integrated injected SO_2 masses. In this work the data base is expanded to the period 1990-2019 and considerably improved for the period 1998-2001. The simultaneous measurements from up to four instruments from 2002 to 2011 enabled us to develop a novel procedure for conversion of aerosol extinction to SO_2 needed for the period before and after ENVISAT.

Our method circumvents problems and uncertainties related to the classical point source approach like dependence on the box size and location, the time interval during which the mass is injected, and effects of microphysical and chemical interactions of SO_2 and sulfate with injected volcanic ash and water in the early phase (Zhu et al., 2020).

Non-eruptive permanently degassing volcanoes represent ~~a smaller~~ another natural source of aerosols, which are treated separately from active explosive volcanic eruptions. For the stratosphere these contribute in most cases only to the background since most of the released SO_2 is removed by oxidation and rainout in the troposphere. This holds also for the medium size eruption of Eyjafjallajökull in 2010 from which almost no SO_2 reached the stratosphere as shown by MIPAS observations. Stratospheric H_2SO_4 is also produced from non-volcanic sulfur precursor gases, like carbonyl sulfide (OCS) (Crutzen, 1976),

dimethyl sulfide (DMS) (Kettle and Andreae, 2000), and tropospheric SO_2 from pollution, ~~constituting a~~ which constitute a source of background concentration of stratospheric aerosol. To improve the accuracy of the sulfur budget of the stratosphere in the CCM, sources and sinks of sulfur as well as its precursor gas components have to be identified.

~~The relevant processes of the stratospheric aerosol life cycle are structured in this paper~~ This paper is structured as follows: section 2 presents satellite data used for entering the volcanic perturbations of aerosols and SO_2 into the model, and for model evaluation. In section 3, the setup used for the climate model simulations is described. Section 4 contains a volcanic sulfur emission inventory with all relevant explosive volcanic eruptions detected between 1990 and 2019, which are included in the model simulations in section 5. The influence of these volcanic eruptions on the stratosphere and climate is analysed in section 6. At the end of section 6 as well as in the final discussion (section 7), the results are discussed in a wider context.

2 Satellite observations

~~We~~ To generate the input data from volcanic eruptions for our simulations, we analysed satellite data sets from two instruments on the European Environmental Satellite (ENVISAT) that was launched on 1 March 2002 and lost signal on 8 April 2012, namely MIPAS and GOMOS. Furthermore, the OSIRIS instrument on board the Odin satellite was used to provide additional data for the period up to 2019. For the period before 2002, we used the SAGE II instrument. ~~Additional SO_2 column data from the Ozone Monitoring Instrument (OMI), Total Ozone Mapping Spectrometer (TOMS), Ozone Mapping and Profiler Suite (OMPS) and other satellites were also used to verify the consistency of the data and to fill in data gaps, marked as white areas in all satellite images~~ The data processing is described in section 4. Some examples for eruptions where simultaneous observations from all these instruments or at least 3 were available for cross validation are presented in the Appendix B.

2.1 Michelson Interferometer for Passive Atmospheric Sounding (MIPAS)

MIPAS was a mid-infrared emission spectrometer on board the ENVISAT satellite. MIPAS scanned the limb, thereby analysing the infrared radiation emitted by the Earth's atmosphere at different tangent altitudes (Fischer et al., 2008).

The atmospheric spectra ranging from $4.15\,\mu$m to $14.6\,\mu$m are inverted to provide vertical profiles of temperature and volume mixing ratios of more than 25 different trace species, like the sulfate aerosol precursor gases SO_2 and OCS (Glatthor et al., 2015, 2017; Höpfner et al., 2013, 2015), as well as H_2O, ozone (O_3), methane (CH_4), nitrous oxide (N_2O), nitrogen dioxide (NO_2) and nitric acid (HNO_3), among others.

Vertical SO_2 profiles (Figure 1) from the MIPAS SO_2 single profile retrieval (Höpfner et al., 2015) were used to identify plumes of volcanic eruptions. We utilised a gridded dataset from these retrievals with a three-dimensional sampling of $60°$ longitude, $10°$ latitude, 1 km altitude with a vertical coverage of $10\,$km to $23\,$km and a temporal averaging of five days. The lower altitude limit varies with the tops of clouds in the troposphere, especially in tropical regions.

The typical estimated random uncertainty for a single measurement of a volume mixing ratio profile is estimated to be ~~.~~ 70–100 pptv. For the gridded dataset used here, systematic uncertainties are more important. These were estimated to be 10–75 pptv (10–180 %) under background concentrations and 10–110 pptv (10–75 %) under volcanic influence (Höpfner et al.,
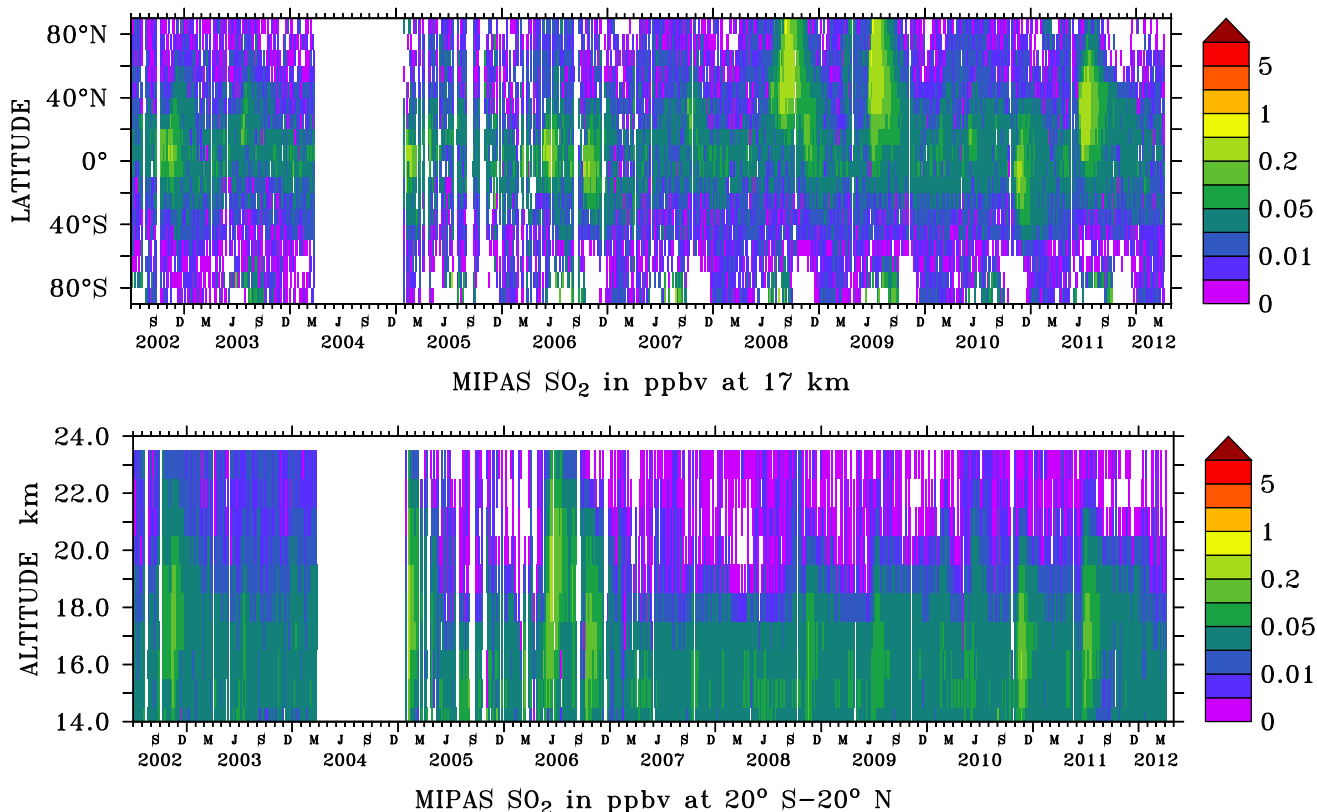
Figure 1. The volume mixing ratios of SO$_2$ in ~~(parts per trillion by volume (10^{-12}))~~ ppbv as derived from the MIPAS instrument (Höpfner et al., 2015). The dataset spans the period 1 July 2002–8 April 2012: Horizontal distribution at 17 km altitude (top) and vertical distribution for tropical regions 20° S–20° N (bottom). White: no data.

2015). ~~Directly after strong volcanic eruptions, the detected mass of SO$_2$ may be underestimated due to saturation of the limb observations as well as the applied cloud clearing scheme (Höpfner et al., 2015).~~

2.2 Global Ozone Monitoring by Occultation of Stars (GOMOS)

The Global Ozone Monitoring by Occultation of Stars (GOMOS) instrument on ENVISAT operates based on the principle of stellar occultation. GOMOS provides data on stratospheric aerosol extinction as well as O$_3$, NO$_2$, nitrogen trioxide (NO$_3$) and air density ~~. The instrument also retrieves the H$_2$O and oxygen (O$_2$) target species in the near-infrared region~~ (Kyrölä et al., 2010). The principle of stellar occultation is described in detail in Bertaux et al. (2010). In short, this self-calibrating sounding method scans the atmosphere by pointing to a star during its sunset or sunrise. The measured spectra vary with the tangent altitude due to the absorption and scattering of light by the different atmospheric species along the line of sight.

In a first step, the GOMOS inversion algorithm determines the slant column density of gaseous species and the slant aerosol optical depth along the optical path (Vanhellemont et al., 2004). This process makes use of reference absorption spectra of

the main absorbing species (such as the ones provided by the MPI-Mainz UV/VIS Spectral Atlas of Gaseous Molecules of Atmospheric Interest (http://satellite.mpic.de/spectral_atlas) and extinction cross-section values representative for aerosols. In a second step, vertical density profiles of the target gas species and vertical profiles of the aerosol extinction coefficient are obtained from the slant quantities (Bertaux et al., 2010).

135   GOMOS uses four spectrometers providing measurements at wavelengths from the UV-visible to the near-IR range in four spectral regions: 248 nm–371 nm, 387 nm–693 nm, 750 nm–776 nm, and 915 nm–956 nm (Robert et al., 2016). As the original inversion algorithm (the operational algorithm IPF) was poorly effective for the retrieval of the aerosol extinction coefficient and only one extinction channel was obtained at the reference wavelength of 550 nm (Vanhellemont et al., 2010), a new retrieval algorithm called AerGOM was designed (Vanhellemont et al., 2016; Robert et al., 2016) in order to improve

140   the spectral inversion. Also, the cross-section spectra were revised using up-to-date reference spectra (Bingen et al., 2019). AerGOM provides the spectral dependence of the aerosol extinction coefficient between about 350 nm and 750 nm.
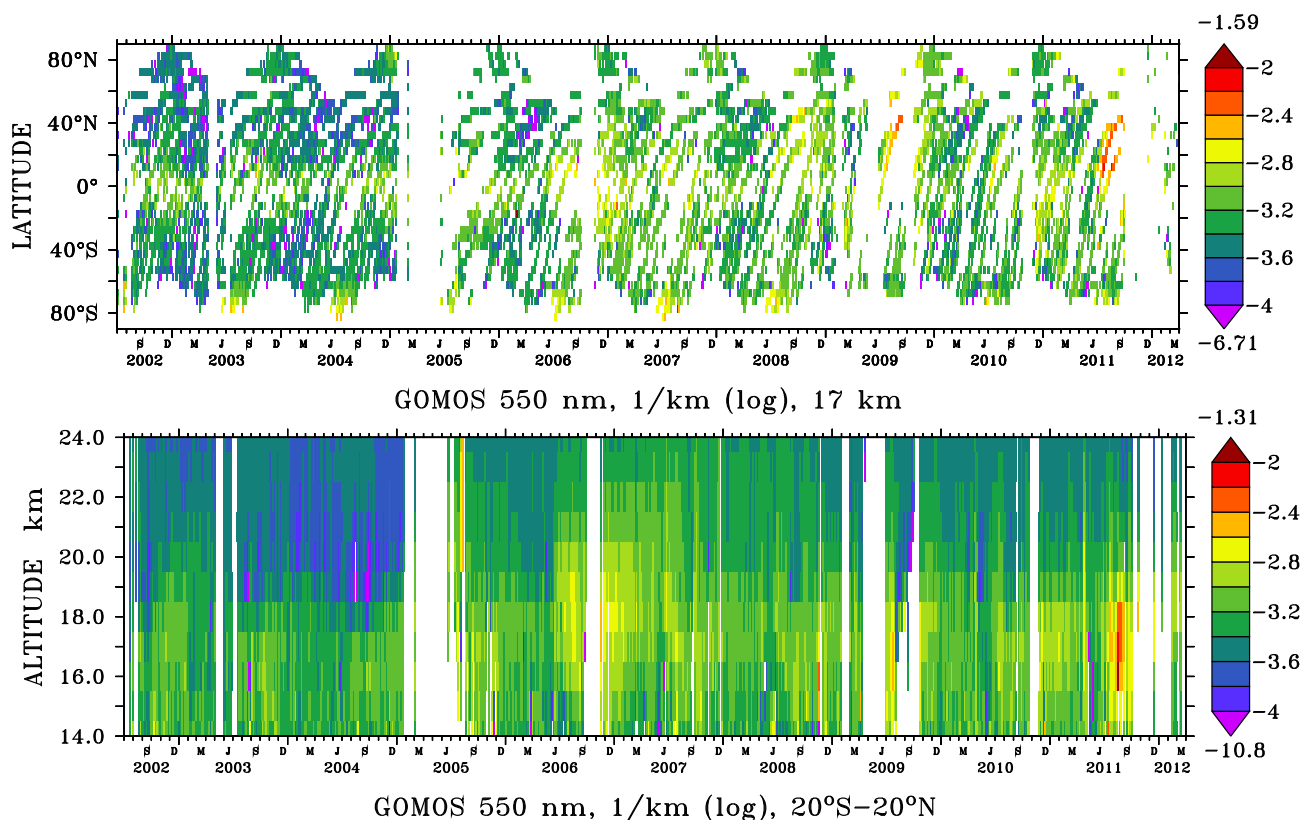
Figure 2. The decadal logarithm (log) of the aerosol extinction coefficient (1/km) as derived from the GOMOS instrument data v.3.00 at **550 nm wavelength** from Bingen et al. (2017). The dataset spans the period 15 April 2002–8 April 2012: Horizontal distribution at 17 km altitude (top) and vertical distribution for tropical regions 20° S–20° N (bottom). Maximum and minimum values appear above (dark red) and below (violet) the colour keys, respectively. White: no data.

The typical extinction uncertainty exhibits large variability in ~~the~~ function of the star parameters (from about 5–15 % in the most favourable cases of bright, hot stars, to about 40–70 % in the less favourable cases of dim, cold stars) (Bingen et al., 2017). A full validation of AerGOM, version 1.0, is presented by Vanhellemont et al. (2016). A main factor influencing the uncertainty is the weakness of the star signal, which is alleviated by the high measurement rate made possible by the abundance of stars. The large variability in magnitude and temperature of the occulted stars also significantly influences the measurement uncertainty (Robert et al., 2016).

From AerGOM, climate data records were processed for use in chemistry-climate models (Bingen et al., 2017), and these are the data sets used in the present study. Figure 2 and Figure 3 ~~illustrates these Level-3 gridded GOMOS aerosol extinction records in version 3.00 of the algorithm, described in Bingen et al. (2017) and shows the~~ show the aerosol extinction from the GOMOS instrument at wavelengths of 550 nm (Figure 2) and ~~.~~ 750 nm (Figure 3), respectively. In both cases, a gridded aerosol extinction dataset is used (CCI-GOMOS dataset in version 3.00, see Bingen et al. (2017)).
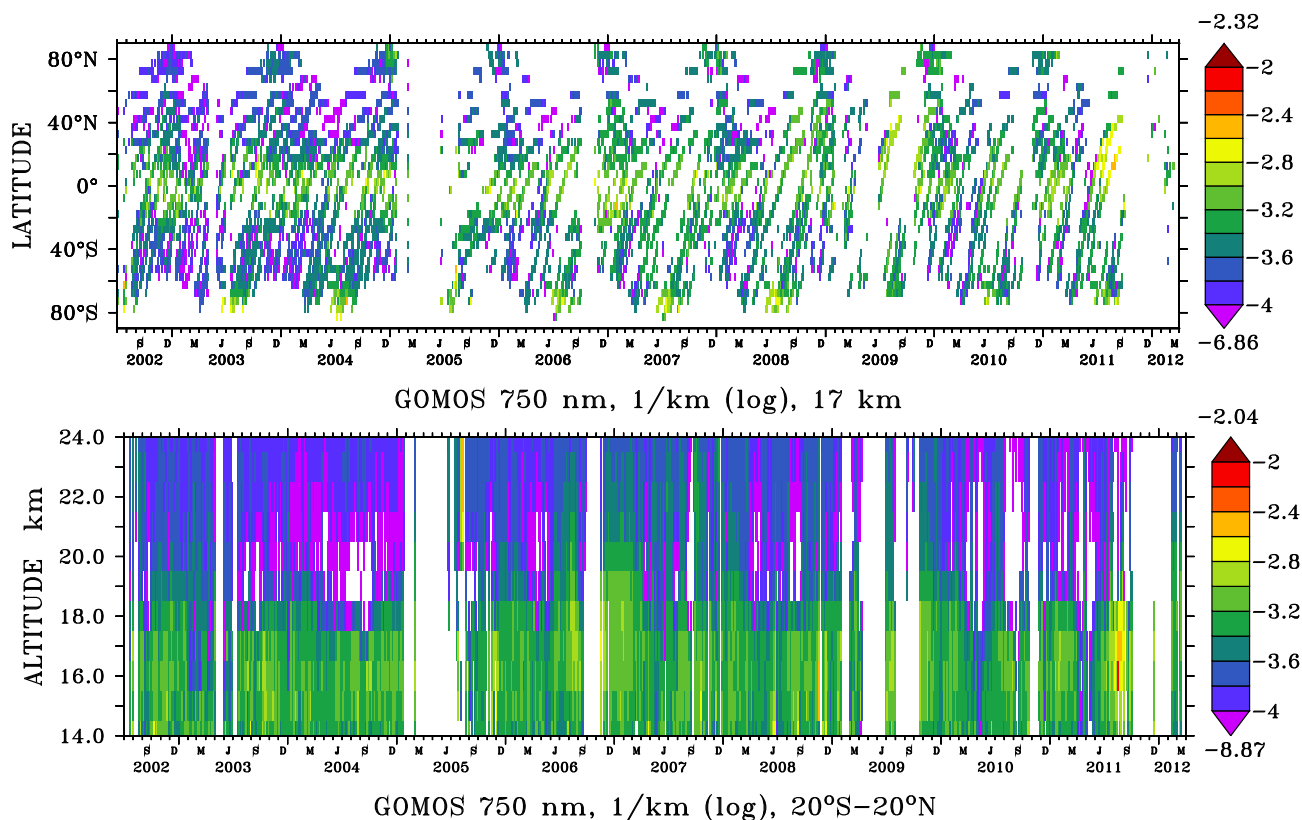
Figure 3. The decadal logarithm (log) of the aerosol extinction coefficient (1/km) as derived from the GOMOS instrument data v.3.00 at **750 nm wavelength** from Bingen et al. (2017). The dataset spans the period 15 April 2002–8 April 2012: Horizontal distribution at 17 km altitude (top) and vertical distribution for tropical regions 20° S–20° N (bottom). Maximum and minimum values appear above (dark red) and below (violet) the colour keys, respectively. White: no data.

The ~~grid resolution~~ resolution of the CCI-GOMOS dataset was optimized to a grid of 5° latitude by 60° longitude and a time resolution of five days. This choice is made possible by the high measurement rate and is more suitable for describing the aerosol distribution than zonal monthly means, because it allows detection of the signature of aerosol patterns with a lifetime of as short as a week (e.g., medium-sized volcanic eruptions). ~~The GOMOS dataset is very important for compensating for data missing from the MIPAS instrument (), where several important eruptions in 2004 and 2006 could not be identified (Bingen et al., 2017).~~

~~The SO$_2$ mixing ratio perturbation is derived from the extinction perturbation observed in a 10-day period beginning about a week after the eruption by dividing by air density, multiplying by a constant and subtracting a typical background.~~

2.3   Optical Spectrograph and InfraRed Imaging System (OSIRIS)

The dataset from the Optical Spectrograph and InfraRed Imaging System (OSIRIS) allowed us to extend the time series beyond April 2012, after which the signal of the ENVISAT satellite was lost. OSIRIS is a limb scatter instrument, which was launched on board the Odin satellite on 20 February 2001 and is still operating today.

OSIRIS performs limb scans of atmospheric radiance spectra at wavelengths from the UV to the near-IR ranges (274 nm–810 nm) (Bourassa et al., 2012). To obtain the vertical profiles of aerosol extinction at altitudes from 10 km to 35 km (Rieger et al., 2015), the aerosol scattering properties are calculated with a refractive index of $1.427 + i7.167 \times 10^{-8}$ using Mie theory at 750 nm wavelength and a sulfate concentration of 75 % H$_2$SO$_4$ and 25 % H$_2$O (Rieger et al., 2018).

For this study, the OSIRIS version 5.10 aerosol retrieval was used until October 2017 and the version 7.1a afterwards (for details see Rieger et al. (2019); Bourassa et al. (2012)). OSIRIS provides a surface coverage from 82° S–82° N, except in polar winter when there is no sunlight and except in the Southern Hemisphere winter when tangent point is not illuminated by the sun (see Figure 4). The vertical grid resolution is 1 km altitude, 5° latitude and 30° longitude with 5-day-averaged time intervals.

The total uncertainty is about 10–15 % in the aerosol layer between 15–30 km, where the sensitivity of the measurements decreases with increasing optical depth. Due to measurement noise, the uncertainty dominates the signal above 30 km and in the troposphere (Rieger et al., 2015). At ~~lower altitudes near~~ altitudes near and below the tropopause, the OSIRIS ~~instrument is more sensitive to cloud perturbation than MIPAS and GOMOS,~~ measurements are sensitive to clouds that may be interpreted as elevated aerosols. This is likely contributing to larger background extinction values measured below approximately 17 km in the tropics, as can be seen in Figure 4 (bottom), and the uncertainty is higher.

~~The SO$_2$ mixing ratio perturbation is derived from the extinction perturbation observed over a 10-day period beginning about a week after the eruption by dividing by air density, multiplying by a constant and subtracting a typical background. If data gaps cause a shift of the time period away from the maximum perturbation or a bias in the zonal average, a correction factor is applied, which can be as high as 5 if the shift is three months as for Calbuco. Correction factors up to 2 have to be applied in some cases because of cloud perturbation or incomplete profiles.~~
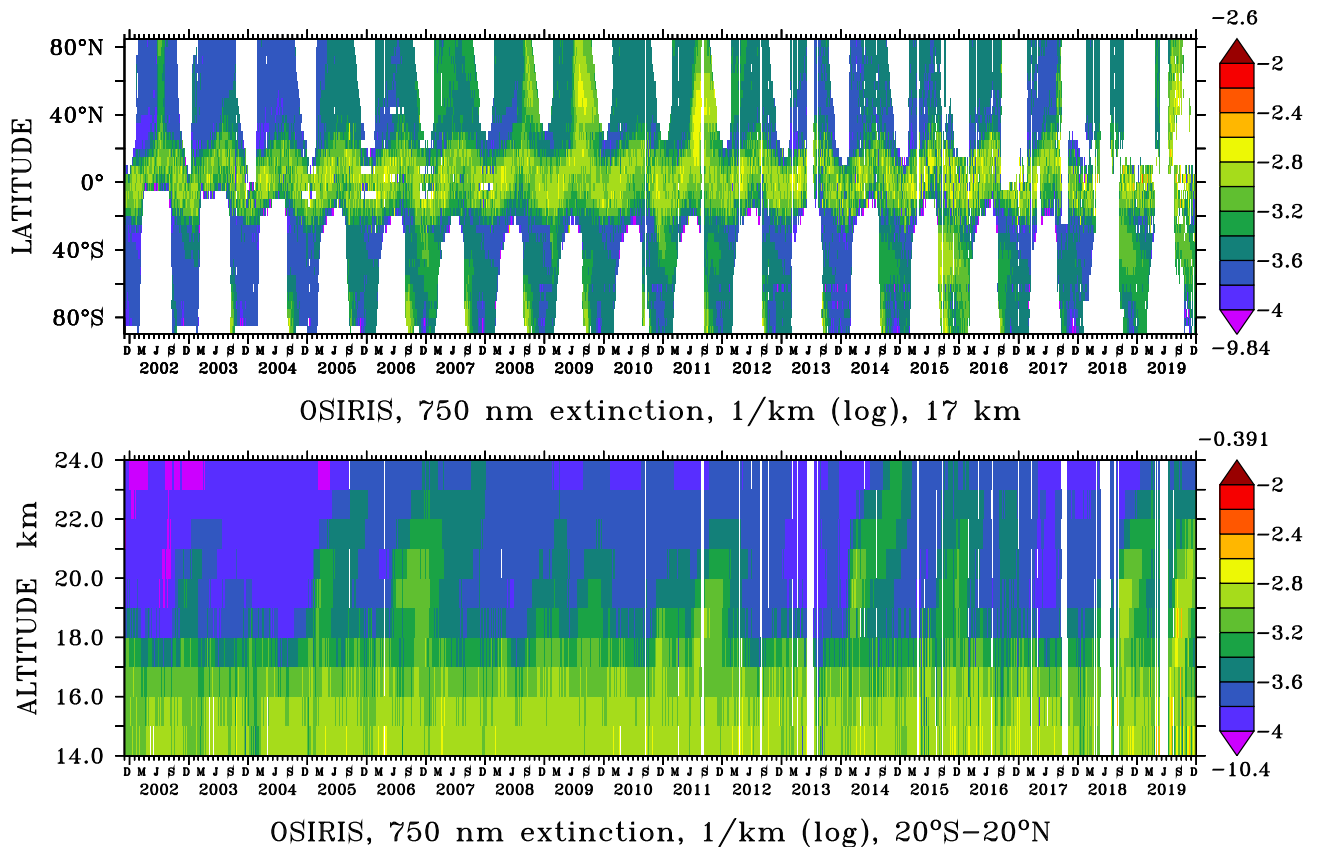
Figure 4. The logarithm log(1/km) of the aerosol extinction as derived from the OSIRIS instrument at **750 nm wavelength** by Bourassa et al. (2012) and Rieger et al. (2019). The dataset spans the period 1 July ~~2002–31~~2002–December ~~December~~ 2019: Horizontal distribution at 16.5 km altitude (top) and vertical distribution for tropical regions 20° S–20° N (bottom). Maximum and minimum values appear above (dark red) and below (violet) the colour keys, respectively. White: no data.

2.4 Stratospheric Aerosol and Gas Experiment II (SAGE II)

185 SAGE II was a solar occultation instrument that performed measurements during sunrise and sunset. The SAGE II aerosol extinction measurements on board the Earth Radiation Budget Satellite (ERBS) started in October 1984 and ended in August 2005. This data set is important for the model setup before the ENVISAT period starting in 2002. The aerosol extinction gridded using 60° longitude and 10° latitude intervals is derived from the V7.00 L2 profiles provided by the Earth Observing System Data and Information System of NASA (EOSDIS) database (Figure 5).

190 The gridded SAGE II dataset used in the present study provides a near-global coverage with latitudes between 80° N to 80° S, with a horizontal grid resolution of 60° in longitude, 10° in latitude and a vertical resolution of 0.5 km between 13 km and 30 km altitude. SAGE II measured in occultation, thus, its measuring principle is similar to that of GOMOS. The two main differences between GOMOS and SAGE II, are that the latter used the sun as light source, which results in a much better signal-
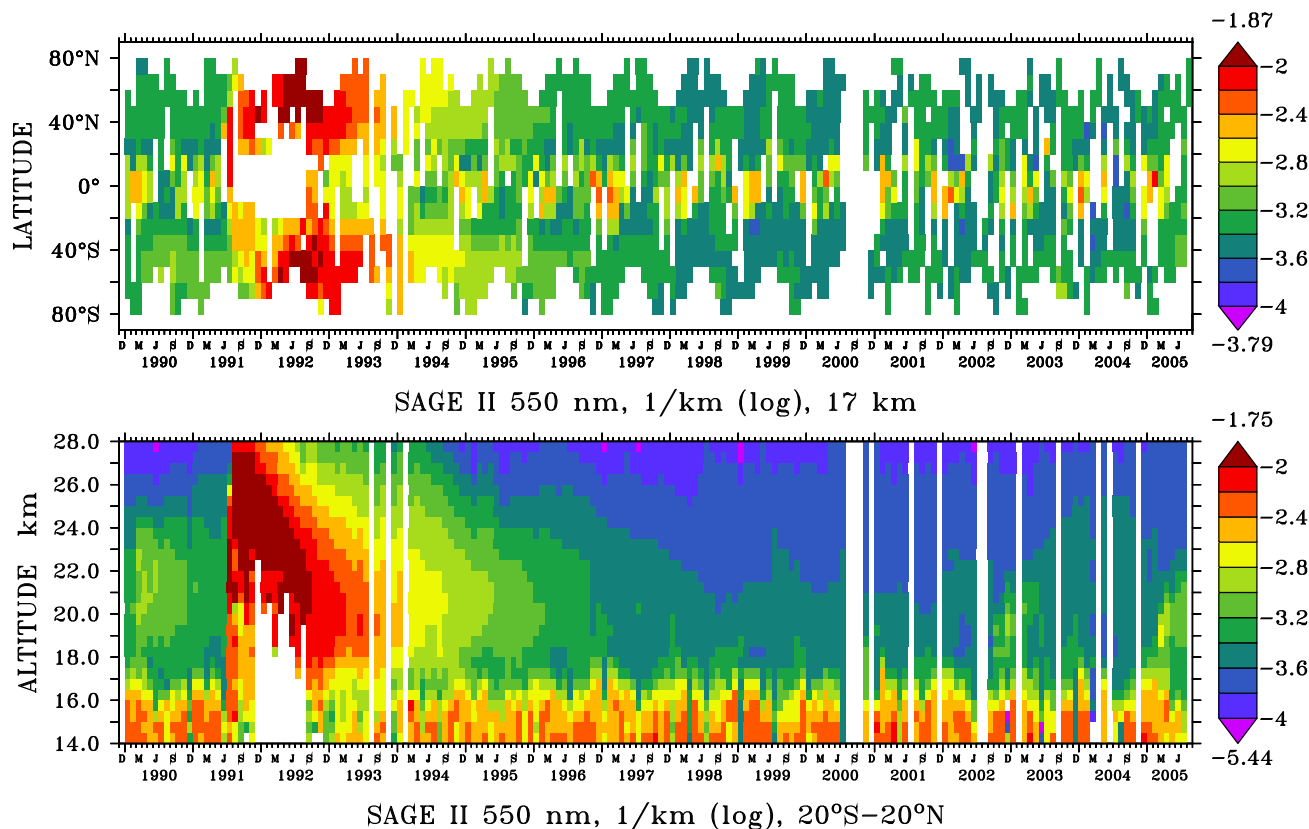
Figure 5. The logarithm of the extinction coefficient (1/km) of the SAGE II instrument from Thomason et al. (2008). The dataset spans the period January 1990–August 2005: Horizontal distribution at 16.75 km altitude (top) and vertical distribution for tropical regions 20° S–20° N (bottom). Maximum and minimum values appear above (dark red) and below (violet) the colour keys, respectively. White: no data.

to-noise ratio. On the other hand, its measurement rate is much lower than that of GOMOS, since only two measurements (one

195   sunrise and one sunset) are possible per orbit, so that a near-global coverage is achieved in about one month.

Measurements occur at seven wavelengths between 386 nm and 1020 nm. The vertical profiles of O_3, NO_2 and water vapor are provided as well as aerosol extinction coefficients at four wavelengths (386, 452, 525 and 1020 nm) from the middle troposphere to the upper stratosphere. ~~An estimate of the aerosol surface area density (SAD) is derived from the extinction coefficients (Thomason et al., 2008). From the SAD, the SO_2 mixing ratio is derived using the parameterisation of~~

200   ~~Grainger et al. (1995).~~

~~The uncertainty of the operational SAD product during background periods is affected by several parameters, including the lack of sensitivity to particles with radii smaller than , the number of degrees of freedom indicated by the averaging kernels of the aerosol extinction at different wavelength channels and the temperature profile used in the data processing (Thomason et al., 2008). The uncertainty also depends by the altitude and is between and , between and and at altitudes above~~

205   ~~(?).~~ After the large eruption of Mount Pinatubo in 1991, "saturation" effects at lower altitudes were observed in the profiles for

more than one year, meaning that the aerosol load was so high that the light signal received by the instrument was below the limit of detection. This effect corresponds to the large white areas for 1991 and 1992 in Figure 5. Red pixels around 14–16 km correspond to measurements contaminated by clouds, increasing the optical depth in the upper troposphere/lower stratosphere (UTLS) region on the lower panel of Figure 5. The perturbations by convective clouds occur mostly over the West Pacific and were excluded in the procedure for estimating the SO_2 injections. The data gaps in the year 2000 were caused by an instrument failure causing SAGE II to be switched off for several months.

The uncertainty of the operational SAD (surface area density) product during background periods is affected by several parameters, including the lack of sensitivity to particles with radii smaller than 100 nm, the number of degrees of freedom indicated by the averaging kernels of the aerosol extinction at different wavelength channels and the temperature profile used in the data processing (Thomason et al., 2008). SAD is used in GLOSSAC.

3 Description of the setup for the EMAC Model

The computations for this study were performed on the Mistral supercomputer at the Deutsches Klimarechenzentrum (DKRZ), Hamburg, Germany. For this purpose, EMAC (ECHAM5/MESSy Atmospheric Chemistry), a coupled atmospheric circulation model consisting of the 5th generation of European Centre Hamburg general circulation model (ECHAM5) and the Modular Earth Submodel System (MESSy) was used .

The model simulations performed in the period from 2002 to 2012 by the global atmospheric chemistry climate model EMAC in Bingen et al. (2017) and Brühl et al. (2018) were extended to 1990 to 2019 in this study.

3.1 The ECHAM5 General Circulation Model

ECHAM5 (5th generation of European Centre Hamburg general circulation model) is an atmospheric general circulation model (Roeckner et al., 2003, 2006), which runs with self-consistent quasi-biennial oscillation (QBO). It is nudged to the meteorological ERA-Interim reanalysis data of the European Centre for Medium-Range Weather Forecasts (ECMWF) up to about For these model simulations, a higher horizontal resolution T63 (1.87° × 1.87°), instead of T42 (2.81° × 2.81°) in Bingen et al. (2017), was chosen. As we here focused focus on the stratosphere, the middle atmosphere version L90 with 90 layers up to 0.01 hPa (∼80 km) and high vertical resolution in the lower stratosphere was needed (Giorgetta et al., 2006). For these model simulations, the horizontal resolution T63 (1.87° × 1.87°) was chosenThe dynamical model is nudged to the meteorological ERA-Interim reanalysis data of the European Centre for Medium-Range Weather Forecasts (ECMWF) up to about 100 hPa.

3.1 The Modular Earth Submodel System (MESSy)

MESSy (Modular Earth Submodel System) is an earth system model , which consists of several submodels (Jöckel et al., 2005, 2006, 2010) . An overview of the submodels used in this study is given in in the appendix.

~~The convection was calculated with the CONVECT submodel (Tost et al., 2006b), with the convection scheme from Tiedtke (1989) and the Nordeng (1994) closure. The convection parametrization is sensitive to the model resolution, which results in differences between different model resolutions in the vertical transport of tracers, like dust and water vapor or ozone (Brühl et al., 2018). To avoid a phase drift, we used the QBO submodel for weak nudging to the QBO zonal wind observations (Giorgetta et al., 2002)~~

240 As EMAC is a very complex chemistry climate model it contains many submodels and functions which are essential for running the simulations but are not directly related to the sulfur cycle, these are mentioned in Appendix A. In this section we focus on the sulfur cycle.

The plumes of outgasing volcanic SO_2 emissions (Diehl et al., 2012) are imported via the OFFEMIS submodel as 3-D field volume emission fluxes (Kerkweg et al., 2006b)~~, while mineral dust emissions are calculated online using the emission~~

245 ~~scheme of Astitha et al. (2012) as part of the ONEMIS submodel~~. The exchange of dimethyl sulfide (DMS) between the air-sea interface of the ocean and the atmosphere is simulated by the AIRSEA submodel (Pozzer et al., 2006).

The gas- and aqueous-phase chemistry in the troposphere and stratosphere is calculated interactively with the CAABA/MECCA (Chemistry As A Boxmodel Application/Module Efficiently Calculating the Chemistry of the Atmosphere) submodel (Sander et al., 2011). Specifically, the chemically generated SO_2 is calculated from fluxes of sulfate precursor gases and further trans-

250 formed to H_2SO_4 (Brühl et al., 2018)~~. The chemistry of the CAABA/MECCA submodel contains photolysis reactions, which need photolysis rate coefficients (J-values) for tropospheric and stratospheric species computed by the JVAL submodel.~~, together with the emitted SO_2 . OH and ozone are fully interactive. CAABA/MECCA is also coupled to the Multiphase Stratospheric Box Model (MSBM) for heterogeneous reactions on aerosols and Polar Stratospheric Clouds (PSCs) (Jöckel et al., 2010) to allow for feedback on ozone. Calculated sulfate is used by MSBM for heterogeneous chemistry. The uptake and

255 oxidation of tracers is considered by the SCAVenging submodel for both liquid and mixed phase clouds (Tost et al., 2006a), also including the aqueous sulfur oxidation of SO_2 to SO_4^{2-} .

For parametrisation of aerosol microphysical processes, we used the Global Modal-aerosol eXtension (GMXe) aerosol module (Kerkweg, 2005; Stier et al., 2005; Vignati et al., 2004) and we described aerosol species using ~~several interacting lognormal aerosol modes, specifically~~ four soluble and three insoluble interacting lognormal aerosol modes. The original

260 mode boundaries of the aerosol size distribution from Pringle et al. (2010) were adapted for this setup to volcanic aerosol conditions in the stratosphere as shown in Table 1 to avoid overly rapid sedimentation of coarse aerosol particles after big volcanic eruptions. The nucleation of new particles consists only of completely soluble sulfate aerosols and is calculated by the parametrisation used by Vehkamäki et al. (2002). Further, the evaporation of liquid sulfate particles back to the gas phase in the middle stratosphere is possible in the model.

265 The ~~nucleation of new particles consists only of completely soluble sulfate aerosols and is calculated by the parametrisation used by Vehkamäki et al. (2002). Further, the evaporation of liquid sulfate particles back to the gas phase in the middle stratosphere is possible in the model. The loss of gas phase species to the aerosol is parametrized in the 3rd EQuilibrium Simplified Aerosol Model (EQSAM3) (Metzger and Lelieveld, 2007). The uptake of gases on wet particles and on acid aerosol particles is included in the model calculation. Calculated sulfate is used by MSBM for heterogeneous chemistry. Furthermore, the~~

270 ~~the~~ AERosol OPTical properties in the model are calculated online with the AEROPT submodel (Dietmüller et al., 2016) and

Mode boundaries	Pringle et al. (2010)	σ	Brühl et al. (2018)	σ
Nucleation mode: soluble	<10 nm	1.69	1 nm–12 nm	1.59
Aitken modes: soluble & insoluble	10 nm–100 nm	1.69	12 nm–140 nm	1.59
Accumulation modes: soluble & insoluble	100 nm–1 μm	1.69	140 nm–3.2 μm	1.49
Coarse modes: soluble & insoluble	>1 μm	2.2	>3.2 μm	1.7

Table 1. Diameters of aerosol mode boundaries in the GMXe submodel for tropospheric (Pringle et al., 2010) and volcanic stratosphere conditions (Brühl et al., 2018), including the corresponding mode distribution width (σ).

are coupled to the GMXe submodel. The resulting extinction coefficient is given at wavelengths of 350, 550, 750 and 1025 nm for comparison with GOMOS, OSIRIS and SAGE. Finally, the ~~parameters from AEROPT~~ aerosol optical properties like wavelength- dependent particle extinction cross section, single scattering albedo, and asymmetry parameter for each aerosol mode from AEROPT (Dietmüller et al., 2016) are used in the radiation scheme as input for the radiative transfer calculations and to calculate the AOD.

The influence of stratospheric aerosol on instantaneous radiative forcing and heating is calculated online for diagnostic purposes ~~.~~ (for details see Dietmüller et al. (2016)). Via multiple calls of the RAD submodel, the instantaneous forcing is simulated with stratospheric aerosol above 100 hPa only and without any aerosol (Brühl et al., 2012). ~~The RAD_FUBRAD sub-submodel is used to calculate the shortwave heating rates from the absorption of UV by O₂ and O₃ in the upper stratosphere and mesosphere. The~~ The lowermost level of the RAD_FUBRAD sub-submodel for the upper atmosphere is shifted from above 70 hPa in the original version of Dietmüller et al. (2016) to 30 hPa–14 hPa to allow for scattering by the aerosol in the simulations with volcanic emissions.

~~Concerning removal mechanisms, the SCAVenging submodel calculates the loss of atmospheric tracers and aerosols by wet deposition, as well as the liquid phase chemistry in clouds and precipitation (Tost et al., 2006a). The uptake and oxidation of tracers is considered for both liquid and mixed phase clouds, also including the aqueous sulfur oxidation of SO₂ to SO₄²⁻.~~

4 ~~Generation of a~~ Volcanic Sulfur Emission Inventory

In a previous inventory of volcanic eruptions based on SO$_2$ vertical profiles from MIPAS, we estimated the aerosol radiative forcing from 2002 to 2011 by simulating the evolution of SO$_2$ in the atmosphere reported by Brühl et al. (2015) and by improving the resulting time series using aerosol measurements from Bingen et al. (2017). The results of these simulations showed that significant discrepancies remained with respect to radiative forcing estimated from measurements (Brühl et al., 2018).

In this work, we further improve the volcanic sulfur emission inventory by analysing additional satellite data sets (section 2) and by including all identified relevant eruptions between 1990 and 2019. ~~The result is presented in and provides the injection time into the model, the coordinates of the ejected plume and the amount of emitted~~ To derive the volcanic 3D SO$_2$ ~~. Each~~

295 ~~volcano is identified by its name if available,~~ perturbation from MIPAS we normally select the five day interval at the time of the eruption and the following one. For medium size eruptions up to 6 consecutive intervals are used to correct for saturation effects or artefacts from the applied cloud clearing scheme in case of ash (Höpfner et al., 2015). As background about 10 pptv, the typical value originating from OCS oxidation, is subtracted. In some cases this value can be larger because of remnants of a previous volcanic event.

300 The amount of sulfur emitted by each single eruption is calculated by integration of SO_2 vertical profiles from MIPAS observations, excluding tropospheric emissions below 12 km at high latitudes, 13 km at mid-latitudes, and 14 km at low latitudes to include possible convective transport from the upper troposphere into the stratosphere in the tropics. The limits in mid and high latitudes above the mean tropopause were selected to exclude cloud perturbations by frontal systems. These limits hold also for the SO_2 estimates from the other instruments.

305 The GOMOS dataset is very important to compensate for data missing from the MIPAS instrument (subsection 2.1), where several important eruptions in 2004 and ~~by the concerned region if the name is unknown. Geological information is received from the "Global Volcanism Program, Smithsonian Institution" ()~~2006 could not be identified (Bingen et al., 2017). An appropriate comparison of SO_2 mixing ratio measurements from MIPAS and of the aerosol extinction from GOMOS requires consideration of a time shift of ~~about~~ one to two weeks as a result of the particle formation from the gas phase. ~~As mentioned~~
310 ~~above, the SAGE II dataset is used to extend the time series to the period preceding ENVISAT's launch, while OSIRIS is used for the period after ENVISAT.~~ We typically select a 10-day period beginning about a week after the eruption. Then the SO_2 mixing ratio perturbation is derived from the extinction by using a ratio between model calculated sulfate volume mixing ratio and its share on extinction in the lower stratosphere of low latitudes, dividing by air density, and subtracting a typical background (see Appendix B). We assume that the spatial patterns of the perturbation of extinction and sulfate are the same as
315 for SO_2. A similar technique is used for OSIRIS and can be used for SAGE II data.

~~The amount of sulfur emitted by each single eruption is calculated by integration of~~ If data gaps cause a shift of the time period away from the maximum perturbation or a bias in the zonal average, a correction factor is applied, which can be as high as 3 if the shift is three months as for Calbuco. To estimate the factor, we iterated calculated extinctions to agree with OSIRIS and also used observations and assumptions by Vernier et al. (2016). Correction factors up to 2 have to be applied in some
320 cases because of data gaps, incomplete profiles or for high latitudes (see Appendix B). On the other hand, the factor can be as small as 0.5 to account for remnants of eruptions occurring 2 weeks before or cloud perturbations.

For SAGE II in most cases the SO_2 mixing ratio is derived using the parameterisation of Grainger et al. (1995) which converts SAD to volume density as first step. We use pressure and temperature provided to convert from mass density to a volume mixing ratio, assuming that observed sulfate is produced from injected SO_2 some weeks ago. With this method it is
325 easier to correct for cloud contamination than by using the extinction directly as above for the other instruments.

Case studies for 3 events, comparing SO_2 ~~vertical profiles from MIPAS observations (Höpfner et al., 2015), excluding tropospheric emissions below at high latitudes, at mid-latitudes, and at low latitudes. The~~ results from the different satellites and the different conversion methods are presented in Appendix B.

Additional SO_2 column data from the Ozone Monitoring Instrument (OMI), Total Ozone Mapping Spectrometer (TOMS),
Ozone Mapping and Profiler Suite (OMPS) (not shown here) and other satellites were also used to verify the consistency of the data and to fill in data gaps, marked as white areas in all satellite images. Especially in 2018 and 2019 the OSIRIS data are so sparse that constraints from instruments like OMPS or analogues events of previous years have to be superimposed for some eruptions.

Geological information and additional observations on plume heights were received from the Global Volcanism Program, Smithsonian Institution (https://volcano.si.edu/). Their reports several times indicate that even VEI 2 eruptions (volcanic explosivity index) can reach the upper troposphere (or lower stratosphere which confirms the satellite observations).

The resulting volcanic emission inventory is presented in Table 2 and provides the injection time into the model, the coordinates of the ejected plume and the amount of emitted SO_2. Each volcano is identified by its name if available, or by the concerned region if the name is unknown. The altitudes and latitudes indicated in the table correspond to the locations of the maximum SO_2 mixing ratios of the volcanic plumes. The longitudes refer to the locations of the volcanoes, because the plumes have been moved by the zonal winds during the time lag between eruption and observation. In the cases of OSIRIS, SAGE and GOMOS this shift can easily be 100 degrees. ~~When comparing the SO_2 emissions reported here with those of Carn et al. (2016), it should be noted that Carn et al. (2016) makes use of total SO_2 emissions, including rapidly removed tropospheric SO_2, while the present study only takes into account the long-lived, climate-relevant stratospheric fraction of the emitted SO_2.~~

It should be noted as well that the date of the volcanic eruption can differ by a few days from the date of injection in the model simulation, because the temporal resolution of the data sets is about five days at least (or weeks in the SAGE period). In ~~some~~ a lot of cases, more than one eruption is found in the same time step within an interval of five days. In such a case, all eruptions are listed in the same line. ~~The integration time~~ Several time the used time period for extraction has to be extended ~~for~~ because of data gaps, which increases the uncertainty and complicates the identification of the right volcano. In such a case, the name of the most probable volcano is tagged with a "?". If the SO_2 emissions of two volcanoes cannot be separated with certainty, both are indicated with a "+" in the same line. This uncertainty is frequent in the Republic of Vanuatu, an island country located in one of the most volcanically active regions in the South Pacific, referred to as "Vanuatu" in Table 2.

When comparing the SO_2 emissions reported here with those of Carn et al. (2016), it should be noted that Carn et al. (2016) makes use of total SO_2 emissions, including rapidly removed tropospheric SO_2, while the present study only takes into account the long-lived, climate-relevant stratospheric fraction of the emitted SO_2. A comparison of the injected volcanic SO_2 masses per year is presented in Appendix C.

Volcano or region	Time	Latitude (°)	Longitude (°)	Altitude (km)	SO_2 (kt)	Instrument
Kelut	11 Feb 1990	-8	112	16, 22	410	S
Gamalama	25 Apr 1990	0	127	16	96	S
Raung (?)	25 Jul 1990	-8	115	16	63	S
Pacaya +	16 Sep 1990	15	-90	16	10	S

Volcano or region	Time	Latitude (°)	Longitude (°)	Altitude (km)	SO$_2$ (kt)	Instrument
Sabancaya	6 Oct 1990	-16	-72	17	75	S
Papua +	12 Jan 1991	-4	145	17	88	S
Fernandina	20 Apr 1991	0	-92	16	118	S
Pinatubo	16 Jun 1991	15	120	23	16942	S
Hudson	10 Aug 1991	-46	-73	18	1276	S
Cerro Negro	10 Apr 1992	12	-87	22	18	S
Spurr	28 Jun 1992	61	-152	17	291	S
Spurr	19 Aug 1992	61	-152	16, 18	298	S
Spurr	18 Sep 1992	61	-152	17	187	S
Lascar	18 Apr 1993	-23	-68	22	376	S
Langila, Galeras (?)	30 Oct 1993	-5, 1	145, -70	17	50	S
Yasur?	17 Mar 1994	-16	165	16	80	S
Rinjani, Nyamuragira, Central America	6 Jul 1994	-8, -1, 12	117, 30, -90	16	63	S
Rabaul	20 Sep 1994	-4	150	18, 22	89	S
Merapi, Ecuador	23 Nov 1994	-7, 1	110, -70	17, 17	48, 57	S
Peru, Africa, Vanuatu	15 Feb 1995	-15, -1, -15	-78, 30, 168	17, 16, 16	7, 43, 25	S
Mexico + Soufriere Hills	10 Aug 1995	16	-98, -62	16	81	S
Peru + Colombia, Rabaul	10 Feb 1996	-15, 5, -4	-80, -80, 150	17, 16, 16	65, 96	S
Soufriere Hills	26 May 1996	16	-62	16	53	S
Soufriere Hills + Mexico, Rabaul	18 Sep 1996	16, -4	-62, -98, 150	16, 16	59, 28	S
Nyamuragira, Manam	3 Dec 1996	-1, -5	30, 145	17, 17	45, 90	S
Manam + Langila	11 Feb 1997	-5	145	17	107	S
Popocatepetl	1 Jul 1997	19	-98	16	32	S
Soufriere Hills, Philippines	20 Oct 1997	16, 16	-62, 121	15, 16	36, 20	S
Soufriere Hills, Papua	26 Dec 1997	16, -8	-62, 150	16, 16	37, 22	S
Tungurahua (?), Vanuatu	2 Feb 1998	-1, -16	-78, 168	17, 16	98, 15	S
Soufriere Hills	4 Jul 1998	16	-62	16	56	S
Manam, Cerro Azul, Nyamuragira	7 Oct 1998	-5, 0, -1	144, -90, 30	17, 17, 16	28, 39, 19	S
Guagua Pinch + Tungurahua, Vanuatu	23 Jan 1999	-1, -16	-78, 165	17, 16	75, 49	S
Cameroon	31 Mar 1999	4	10	16	63	S
Mayon, Colombia	22 Jun 1999	13, 2	124, -80	17, 16	41, 46	S
Soufriere Hills +	24 Jul 1999	16	-62	17	42	S
Ulawun, Tungurahua + Guagua Pichincha	16 Nov 1999	-5, -1	150, -78	17	31, 51	S
Vanuatu, Nyamuragira, Tungurahua	4 Feb 2000	-16, -1, 0	165, 30, -78	17, 16, 16	33, 41, 12	S
Mayon + Vanuatu, Tungurahua	29 Feb 2000	13, -16, -1	124, 168, -78	16, 16, 16	25, 32	S

Volcano or region	Time	Latitude (°)	Longitude (°)	Altitude (km)	SO$_2$ (kt)	Instrument
Ulawun (+ Miyakejima)	26 Sep 2000	-5	150	17	42	S
Nyamuragira, Mayon(?)	13 Feb 2001	-1, 13	30, 124	16, 18	47, 88	S
Ulawun	29 Apr 2001	-5	150	16	41	S
Mayon, Lopevi	23 Jun 2001	13, -16	124, 168	16, 16	49, 22	S
Tungurahua, Soufriere Hills	7 Aug 2001	0, 16	-78, -62	16, 16	29, 46	S
Africa, Tungurahua +	25 Sep 2001	-1, 0	30, -78	16, 16	31, 47	S
Tungurahua (+ Manam), Nyiragongo	14 Jan 2002	-1(-4), -1	-78(144), 30	17, 15	83, 19	S
Tungurahua (+ Africa)	20 Mar 2002	-1	-78 (30)	17	77	S
Nyamuragira	23 Jul 2002	-1	30	15	23	M
Witori	2 Aug 2002	-6	150	14	18	M
Ruang	26 Sep 2002	2	125	18	71	M, G
El Reventador	5 Nov 2002	0	-78	17	77	M, G
Nyiragongo, Lokon	9 Jan 2003	-1, 1	30, 125	15, 16	12, 10	M, G
Nyiargongo, Lokon (Rabaul?)	5 Mar 2003	-5, 1	30, 125	17, 15	12, 13	M, G
Anatahan, Nyiaragongo, Ulawun	14 May 2003	16, -1, -5	143, 30, 150	16, 16, 17	9, 15, 6	M
Lewotobi, Kanlaon	13 Jun 2003	-8, 10	123, 123	15, 15	9, 15	M, G
Soufriere Hills	13 Jul 2003	16	-62	17	41	M, G
Gamalama, Japan	17 Aug 2003	1, 33	128, 131	16, 16	8, 7	M, G
Bezymianny or Klyuchevskoy	6 Sep 2003	56	160	14	8	G
Lokon, Soufriere Hills + Masaya	26 Sep 2003	2, 15	125, -62	16, 16	7, 5	M, G
Rabaul	10 Nov 2003	-5	150	16	17	M, G
Rabaul	5 Dec 2003	-5	150	16	13	M, G
Rabaul, Nyiaragongo?	9 Jan 2004	-5, -1	150, 30	17, 15	11, 9	M, G
Langila, Nyiaragongo?	3 Feb 2004	-5, -1	150, 30	17, 17	11, 3	M, G
Soufriere Hills	4 Mar 2004	10	-62	17	22	M, G
Nyamuragira, Awu + Tengger	12 Jun 2004	-1, 4, -8	30, 125, 112	17, 15	20, 18	G
Pacaya, Galeras	17 Jul 2004	15, 1	-91, -77	17, 17	11, 11	G
Galeras	11 Aug 2004	1	-77	16	15	G
Vanuatu, Rinjani + Kerinci	30 Sep 2004	-16, -8, -2	168, 116, 101	15, 15, 17	7, 15	G
Manam, Soputan	30 Oct 2004	-4, 1	144, 125	16, 16	8, 11	G
Manam, Nyiragongo	24 Nov 2004	-4, -1	144, 30	17, 15	18, 11	G
Nyiaragongo, Reventador	4 Dec 2004	0, 0	30, -77	16, 16	19, 5	G
Vanuatu, Soputan	24 Dec 2004	-16, 1	168, 125	17, 15	15, 16	G
Manam	28 Jan 2005	-4	144	18	130	M, G
Anatahan, (+)	3 Apr 2005	16	143	15	15	M
Anatahan, Soufriere Hills	23 Apr 2005	16, 16	143, -62	16, 16	21, 21	M

Volcano or region	Time	Latitude (°)	Longitude (°)	Altitude (km)	SO$_2$ (kt)	Instrument
Anatahan, Fernadina, Vanuatu	18 May 2005	16, 0, -16	143, -91, 168	15, 15, 15	8, 11, 6	M
Anatahan, Santa Ana	12 Jun 2005	16, 14	143, -90	15, 15	12, 9	M
Anatahan, Soufriere Hills	12 Jul 2005	16, 16	143, -62	15, 15	14, 10	M
Anatahan, Raung	6 Aug 2005	16, -8	143, 113	15, 15	13, 20	M
Anatahan, Raung	16 Aug 2005	16, -8	143, 113	15, 15	14, 17	M, G
Santa Ana	5 Oct 2005	14	-90	17	32	M
Sierra Negra, Dabbahu	25 Oct 2005	-1, -13	-91, 40	15, 15	16, 22	G
Karthala, Galeras	24 Nov 2005	-10, -2	43, -80	16, 16	13, 11	M, G
Soputan, Lopevi	24 Dec 2005	1, -16	125, 168	16, 16	23, 13	M, G
Rabaul +	23 Jan 2006	-5	152	16	25	G
Manam, Chile	4 Mar 2006	-5, -40	144, -70	17, 16	58, 6	G, T
Cleveland	14 Mar 2006	53	-170	13	8	G
Ecuador, Tinakula, Lascar	18 Apr 2006	-5, -10, -23	-78, 166, -68	17, 17, 17	13, 17, 3	M
Soufriere Hills	23 May 2006	16	-62	19	125	M, G, T
Kanlaon	2 Jul 2006	10	123	20	42	M
Tungurahua, Rabaul	16 Aug 2006	-2, -4	-78, 150	19, 17	40, 20	M, G, T
Rabaul	10 Oct 2006	-4	150	17	131	M, T
Ubinas, Vanuatu	25 Oct 2006	-20, -20	-70, 168	17, 15	8, 25	M
Ambrym	9 Nov 2006	-10	160	17	27	M, T
Nyamuragira, Mexico	29 Nov 2006	5, 5	30, -90	17, 15	28, 21	M, G, T
Bulusan, Soputan, Vanuatu	24 Dec 2006	13, 1, -16	125, 125, 168	18, 16, 15	8, 8, 14	M, G
Karthala, Bulusan, Lascar, Shiveluch, Vanuatu	23 Jan 2007	-10, 13, -23, 57, -16	43, 125, -68, 160, 168	17, 17, 15, 15, 15	5, 5, 6, 7, 5	M, G, T
Nevado del Huila, Karthala, Vanuatu	22 Feb 2007	0, -10, -16	-70, 43, 168	16, 15, 16	8, 10, 8	M, G, T
Etna, Reventador, Ambrym	24 Mar 2007	38, 0, -16	15, -78, 160	15, 16, 17	8, 17, 14	M, G, T
Piton de la Fournaise, Reventador +	8 Apr 2007	-20, 0	57, -80	16, 16	22, 11	M, G, T
Ulawun, Vanuatu, Nevado del Huila	3 May 2007	-5, -25, 3	150, 160, -70	15, 15, 15	11, 5, 6	M, G, T
Papua, Kamchatka, Nyamuragira, Ubinas + Lascar	13 May 2007	-10, 50, 0, -20	150, 150, 30, -75	16, 16, 16, 16	6, 1, 10, 6	M, G
Llaima, Vanuatu, Bulusan	23 May 2007	-30, -15, 13	-70, 160, 125	18, 15, 17	10, 6, 7	M, G
Soputan, Bezymianny, Telica	12 Jun 2007	1, 56, 13	125, 160, -87	16, 14, 15	13, 7, 9	M, G
Lengai, Mexico	2 Jul 2007	2, 20	29, -90	16, 15	14, 9	M
Raung, Japan (+)	27 Jul 2007	-5, 35	110, 130	15, 15	10, 10	M
Manda Hararo, Java	11 Aug 2007	12, -5	40, 115	17, 15	13, 14	M, T
Vanuatu, Mexico	20 Sep 2007	-5, 20	180, -90	16, 16	8, 13	M
Jebel al Tair, Galeras	5 Oct 2007	16, 1	42, -80	16, 16	41, 8	M, T

Volcano or region	Time	Latitude (°)	Longitude (°)	Altitude (km)	SO$_2$ (kt)	Instrument
Galeras, Jebel al Tair, Soputan	4 Nov 2007	-2, 15, -5	-80, 42, 110	16, 16, 16	7, 5, 8	M, G
Soputan or Krakatau, Galeras, Chikurachki	14 Nov 2007	-5, -1, 50	110, -75, 155	16, 16, 15	9, 8, 10	M
Talang, Galeras	9 Dec 2007	0, 0	100, -75	16, 16	10, 12	M
Ulawun?	19 Dec 2007	1	150	17	17	M, G
Nevado del Huila, Llaima	3 Jan 2008	1, -35	-75, -71	17, 15	26, 4	M
Galeras, Anatahan	23 Jan 2008	-3, 15	-80, 145	16, 16	14, 7	M
Tungurahua, Papua	12 Feb 2008	-5, -5	-80, 155	16, 17	13, 10	M
Batu Tara (+)	13 Mar 2008	-5	125	16	26	M, G
Lengai, Andes, Kerinic	28 Mar 2008	-5, 5, -2	36, -80, 101	16, 16, 16	6, 4, 7	M
Egon, Nevado del Huila	12 Apr 2008	-5, 5	122, -76	15, 17	14, 9	M
Mexico, Ibu, Chaiten	27 Apr 2008	15, 1, -35	-90, 125, -70	16, 16, 16	9, 11, 3	M
Mexico, Barren Island, Chaiten	12 May 2008	10, 10, -35	-90, 90, -70	14, 16, 14	10, 14, 5	M
Soputan, Nicaragua/Costa Rica	16 Jun 2008	1, 1	125, -85	16, 16	26, 8	M
Okmok, Soputan	21 Jul 2008	53, 1	-168, 125	16, 16	51, 27	M
Kasatochi	15 Aug 2008	52	-175	17	273	M, G
Dallafilla, Nevado del Huila, Reventador	13 Nov 2008	14, 3	40, -78	17, 17	39, 28	M
Karangetang, Galeras, Japan	18 Dec 2008	3, 0, 30	125, -80, 130	17, 17, 15	15, 10, 9	M, G
Barren Island, Galeras	2 Jan 2009	10, 3	90, -80	17, 15	10, 10	M
Indonesia?, Galeras	27 Jan 2009	-5, 0	100, -80	16, 16	12, 10	M
Galeras, Villarrica, Karangetang, Vanuatu	16 Feb 2009	-2, -35, 3, -16	-78, -75, 100, 168	16, 15, 16, 17	11, 6, 6, 7	M
Redoubt, Galeras	28 Mar 2009	60, 0	-155, -75	13, 15	61, 43	M
Fernandina, Nyamuragira	12 Apr 2009	0, 0	-90, 30	16, 16	12, 16	M
Galeras + Reventador	7 May 2009	0	-75	15	25	M
Rinjani, Vanuatu, Reventador	22 May 2009	-5, -15, 3	116, 165, -80	16, 16, 16	4, 4, 13	M
Sarychev, Manda Hararo	21 Jun 2009	48, 12	153, 40	16, 16	446, 82	M, G
Vanuatu, Mayon, Galeras	4 Oct 2009	-15, 13, 2	165, 120, -80	17, 17, 17	4, 6, 10	M
Tungurahua, Hawaii, Vanuatu	19 Oct 2009	5, 20, - 16	-76, -155, 165	16, 16, 16	7, 5, 5	M, G
Galeras, Karkar, Vanuatu	3 Dec 2009	0, -5, -16	-78, 146, 165	17, 17, 17	12, 10, 4	M
Mayon, Nyamuragira, Vanuatu	2 Jan 2010	13, 0, -15	120, 30, 168	16, 16, 16	8, 8, 9	M
Turrialba, Vanuatu	17 Jan 2010	5, -15	-82, 168	16, 16	9, 9	M
Soufriere Hills	16 Feb 2010	16	-62	17	36	M
Arenal, Indonesia, Vanuatu	2 Apr 2010	9, 0, -16	-84, 120, 168	15, 15, 15	14, 12, 5	M
Tungurahua, Dukono, Vanuatu	2 May 2010	-5, 2, -16	-78, 128, 168	16, 16, 16	14, 10, 7	M
Pacaya, Ulawun, Sarigan	6 Jun 2010	15, -5, 16	-91, 150, 145	17, 16, 15	27, 6, 4	M
Ulawun, Costa Rica, Miyakejima	16 Jul 2010	-5, 15, 35	150, -87, 140	16, 16, 16	8, 13, 6	M, G
Karangetang, Nicaragua, Vanuatu	15 Aug 2010	3, 15, -16	125, -85, 168	16, 16, 16	12, 12, 6	M

Volcano or region	Time	Latitude (°)	Longitude (°)	Altitude (km)	SO$_2$ (kt)	Instrument
Galeras, Sinabung	30 Aug 2010	5, 5	-77, 100	16, 16	10, 12	M
Karangetang, Barren Island	4 Oct 2010	3, 12	125, 94	16, 16	20, 13	M
Merapi	8 Nov 2010	-7	110	17	97	M
Tengger, Tungurahua, Chile	23 Dec 2010	-8, -3, -40	110, -78, -75	17, 17, 17	16, 13, 8	M
Tengger	7 Jan 2011	-8	110	16	24	M
Lokon, Planchon, Bulusan	26 Feb 2011	1, -35, 13	125, -75, 125	16, 15, 16	13, 4, 12	M
Karangrtang, Sangay, Planchon	23 Mar 2011	2, -2, -35	125, -78, -75	15, 15, 15	10, 10, 5	M
Galeras?, Karangetang	12 Apr 2011	5, 5	-77, 128	16, 16	10, 9	M
Tungurahua, Dukono, Vanuatu	2 May 2011	2, 2, -16	-78, 128, 160	16, 16, 15	13, 9, 5	M
Grimsvötn, Lokon	27 May 2011	65, 1	-20, 125	14, 16	18, 27	M
Puyehue	11 Jun 2011	-41	-71	13	23	G
Nabro	21 Jun 2011	13	41	18	406	M, G
Soputan, Marapi	20 Aug 2011	1, 0	125, 100	18, 16	9, 3	M, G
Manam, Tungurahua	19 Oct 2011	-4, -3	144, -78	16, 16	8, 8	M
Nyamuragira	18 Nov 2011	-2	29	16	31	M
Gamalama, Nyamuragira	18 Dec 2011	1, -1	128, 29	16, 15	19, 13	M
Vanuatu, Nyamuragira	12 Jan 2012	-16, -1	168, 29	16, 14	14, 12	M
Vanuatu, Nyamuragira	11 Feb 2012	-16, -1	168, 29	17, 17	16, 15	M
Nevado del Ruiz, Marapi	12 Mar 2012	-3, 0	-76, 100	16, 17	12, 15	M
Nyamuragira, Mexico	7 Jun 2012	-1, 20	29, -95	16, 15	30, 4	O
Soputan, Nevado del Ruiz, Mexico	27 Aug 2012	1, 5, 20	124,-76,-95	16, 16, 15	30, 15, 5	O
Nyamuragira, Mexico, Peru	14 Oct 2012	-1, 20, -20	29, -95, -70	16, 16, 15	40, 15, 10	O
Nyamuragira, Paluweh, Nevado del Ruizz	7 Nov 2012	-1, -8, 5	29, 122, -76	15, 16, 17	20, 30, 17	O
Copahue, Lokon +	22 Dec 2012	-38, 1	-71, 125	15, 17	10, 45	O
Paluweh, Karkar	3 Feb 2013	-8, -5	122, 145	16, 17	25, 22	O
Karkar, Vanuatu (+?)	10 Mar 2013	-5, -16	145, 168	17, 16	24, 20	O
Rabaul, Nevado del Ruiz, Nyamuragira	18 Apr 2013	-3, 5, -1	150,-76, 29	17, 17, 16	40, 9, 20	O
Mayon, Turrialba, Pavlof	8 May 2013	13, 10, 55	124, -84, -162	17, 16, 14	35, 24, 6	O
Rabaul, Mexico	10 Jul 2013	-3, 20	150, -95	16, 15	30, 15	O
Pacaya	15 Aug 2013	15	-91	16	43	O
Sinabung, Ubinas	15 Sep 2013	3, -16	98, -71	17, 15	35, 8	O
Merapi, Nyamuragira, Pacaya	18 Nov 2013	-7, -1, 15	110, 29, -91	17, 17, 15	30, 13, 8	O
Sinabung, Nyamuragira	9 Dec 2013	3, -1	98, 29	17, 16	26, 15	O
Sinabung +	11 Jan 2014	3	98	16	29	O
Kelut	15 Feb 2014	-8	112	20	170	O
Merapi, Tungurahua	27 Mar 2014	-7, -1	110, -78	16, 16	31, 33	O

Volcano or region	Time	Latitude (°)	Longitude (°)	Altitude (km)	SO$_2$ (kt)	Instrument
Santa Maria, Semeru	9 May 2014	15, -8	-91, 113	16, 16	25, 39	O
Sangeaang-Api	31 May 2014	-8	119	17	60	O
Nyamuragira, Pavlof, Fuego, Dukono (Tungurahua)	9 Jul 2014	-1, 55, 14, 2	29, -162, -91, 128	16, 15, 15, 16	20, 10, 12, 20	O
Rabaul, Fuego	29 Aug 2014	-3, 14	150, -91	16, 16	36, 20	O
Nyamuragira	11 Sep 2014	-1	29	15	30	O
Ontakesan	27 Sep 2014	36	137	17	34	O
Sinabung, Turrialba	23 Oct 2014	3, 10	98, -84	17, 16	34, 17	O
Fogo, Semeru, Ubinas	24 Nov 2014	15, -8, -16	-24, 113, -71	17, 17, 16	11, 33, 11	O
Nevado del Ruiz, Nyamuragira, Vanuatu	16 Dec 2014	5, -1, -16	-76, 29, 168	15, 17, 16	8, 12, 21	O
Nyamuragira, Vanuatu, Honga Tonga	14 Jan 2015	-1, -16, -21	29, 168, -175	16, 16, 15	21, 17, 13	O
Vanuatu, Nyamuragira, Soputan	16 Feb 2015	-16, -1, 1	168, 29, 124	17, 16, 16	13, 13, 13	O
Soputan, Nevado del Ruiz, Santa Maria, Villarrica	8 Mar 2015	1, 5, 15, -39	125,-76,-91,-72	17, 16, 15, 15	14, 14, 8, 5	O
Tungurahua?, Batu Tara?	5 Apr 2015	-1, -8	-78, 124	17, 17	17, 22	O
Calbuco	25 Apr 2015	-41	-73	18	292	O
Manam, Tungurahua?	8 May 2015	-4, -1	144, -78	17, 17	24, 25	O
Wolf, Aira + Kuchinoerabujima	26 May 2015	0, 32, 30	-91, 131, 130	16, 15	63, 20	O
Raung	4 Jul 2015	-5	110	17	27	O
Cotopaxi, Raung, Suwanosjima, Manam	14 Aug 2015	0, -5, 30, -4	-80,110,130,144	16, 16, 16, 20	24,18,10,16	O
Nev. Ruiz + Reventador, Fuego, Sumatra	21 Sep 2015	5, 14, 3	-76, -91, 98	16, 17, 16	13, 8, 19	O
Sinabung, Fuego, Cotopaxi, Copahue	15 Oct 2015	3, 14, 0, -38	98, -91, -80, -71	16, 17, 15, 15	30, 15, 6, 13	O
Lascar, Sinabung, Nyamuragira, Fuego	30 Oct 2015	-23, 3, -1, 14	-70, 98, 29, -91	17, 17, 16, 16	13,17,12,17	O
Vanuatu, Tungurahua, Telica, Rinjani	17 Nov 2015	-16, -1,13, -5	168,-78,-87,116	18, 17, 17, 16	18,20,10,18	O
Vanuatu, Reventador, Tengger	5 Dec 2015	-16, 0, 2	168, -78, 120	17, 16, 16	16, 15, 12	O
Reventador, Sinabung	18 Dec 2015	0, 3	-78, 100	17, 16	16, 16	O
Soputan +, Reventador, Fuego	8 Jan 2016	1, 0, 14	125, -78, -91	16, 17, 14	25, 19, 5	O
Semeru, Fuego	10 Feb 2016	-8, 14	113, -91	17, 16	34, 25	O
Vanuatu +, Tungurahua	27 Feb 2016	-16, -1	168, -78	16, 16	24, 16	O
Tungurahua, Sinabung +, Pavlof	15 Mar 2016	-1, 3, 55	-78, 98, -162	16, 17, 15	23, 26, 7	O
Reventador, Sinabung +, Fuego, Aira	13 Apr 2016	0, 3, 14, 32	-78, 98, -91, 131	17, 16, 15, 15	18, 30, 17, 6	O
Fuego, Nyamuragira + Ecuador, Langila, Sinabung	7 May 2016	14, -1, -5, 3	-91, 29, 150, 98	16, 17, 16, 17	16, 18, 16, 26	O
Bulusan, Sinabung, Semeru, Mexico	10 Jun 2016	13, 3, 8, 15	125,98,113,-100	17, 16, 17, 16	16,14,16,10	O
Rinjani, Sinabung, Santa Maria	1 Aug 2016	-5, 3, 15	116, 98, -91	16, 16, 16	10, 30, 24	O
Sinabung + Vanuatu, Fuego	28 Aug 2016	-16, 14	168, -91	16, 16	42, 23	O

Volcano or region	Time	Latitude (°)	Longitude (°)	Altitude (km)	SO$_2$ (kt)	Instrument
Ubinas, Sinabung	3 Oct 2016	-16, 3	-71, 98	15, 16	16, 26	O
Sabancaya, Sinabung + Bulusan	5 Nov 2016	-16, 3	-72, 98	16, 16	38, 46	O
Dukono, Vanuatu, Sabancaya	12 Dec 2016	2, -16, -16	128, 168, -72	17, 18, 15	30, 28, 28	O
Sabancaya, Reventador, Sinabung + Vanuatu	10 Jan 2017	-16, 0, 3	-72, -78, 98	16, 17, 17	20, 30, 23	O
Sabancaya, Colima, Sinabung	4 Feb 2017	-16, 19, 3	-72, -104, 98	15, 16, 16	17, 15, 25	O
Sabancaya, Dukono, Fuego, Manam + Vanuatu, Bogoslof, Nevados de Chillán	5 Mar 2017	-16, 2, 14, -16, 53, -37	-72, 128, -91, 168, -170, -71	16, 17, 17, 17, 15, 15	10, 18, 8, 28, 4, 5	O
Sabancaya, Nevado del Ruiz, Sinabung, Vanuatu, Klyuchevskoy	10 Apr 2017	-16, 5, 3, -16, 56	-72, -75, 98, 168, 160	16, 16, 16, 16, 15	8, 15, 19, 17, 2	O
Sinabung, Manam, Fuego	5 May 2017	3, -4, 14	98, 145, -91	16, 17, 17	26, 10, 19	O
Sheveluch + Bogoslof	19 May 2017	57	161	15	20	O
Santa Maria, Sheveluch +, Manam	16 Jun 2017	15, 57, -4	-91, 161, 145	16, 16, 15	11, 33, 6	O
Fuego, Sinabung +, Sheveluch +	5 Jul 2017	14, 3, 57	-91, 98, 161	15, 16, 15	22, 21, 4	O
Sinabung, Cristobal + Fuego, Sheveluch + Bogoslof	8 Aug 2017	3, 13, 54	98, -87, -168	16, 17, 16 (26?)	31, 27, 5	O
Tinakula, Ambae	21 Oct 2017	-10, -15	166, 168	15, 15	60	O
Agung, Ambae, Sabancaya	27 Nov 2017	-8, -15, -5	116, 168, -80	15, 16, 15	22, 7, 12	O
Mayon, Vanuatu, Sabancaya	22 Jan 2018	13, -15, -5	124, 168, -80	15, 17, 16	7, 20, 16	O
Fuego, Vanuatu	1 Feb 2018	14, -15	-91, 168	16, 16	20, 17	O
Sinabung, Vanuatu	19 Feb 2018	3, -15	98, 168	16, 16	14, 21	O
Ambae, Vanuatu	26 Mar 2018	-15	168	16	60	O
Ambae	6 Apr 2018	-15	168	17	91	O
Sabancaya	15 May 2018	-16	-72	16	16	O, T
Fuego	3 Jun 2018	14	-91	16	15	O, T
Fernandina	17 Jun 2018	0	-92	15	8	T
Agung, Sabancaya	28 Jun 2018	-8, -16	115, -72	17, 16	33, 23	O, T
Sierra Negra	8 Jul 2018	-1	-92	15	25	T
Ambae	20 Jul 2018	-15	168	17	228	O, T
Manam, Sabancaya	25 Aug 2018	-3, -16	144, -72	17, 16	25, 12	O, T
Krakatau, Sabancaya	23 Sep 2018	-6, -16	105, -72	16, 16	5, 11	O
Manam, Soputan, Reventador + Sangay	4 Oct 2018	-3, 1, 0	144, 125, -78	16, 16, 16	7, 4, 22	O
Nev.Ruiz, Sabancaya	24 Oct 2018	5, -16	-75, -72	16, 16	22, 11	O
Fuego, Sabancaya, Krakatau	6 Nov 2018	14, -16, -6	-91, -72, 105	16, 16, 15	10, 16, 19	O
Fuego, Sabancaya, Bagana	26 Nov 2018	14, -16, -6	-91, -72, 155	16, 16, 16	8, 9, 12	O
Sabancaya, Manam, Soputan, Vanuatu	8 Dec 2018	-16, -3, 1,-16	-72,144,125,168	16, 17, 16, 15	24, 8, 4, 6	O

Volcano or region	Time	Latitude (°)	Longitude (°)	Altitude (km)	SO$_2$ (kt)	Instrument
Krakatau, Vanuatu, Sabancaya	23 Dec 2018	-6, -16, -16	105, 168, -72	16, 15, 16	7, 6, 20	O
Krakatau, Sabancay, Manam	4 Jan 2019	-6, -16, -3	105, -72, 144	17, 17, 16	5, 20, 9	O
Manam, Sabancaya	24 Jan 2019	-3, -16	144, -72	17, 16	23, 14	O
Manam, Sabancaya	14 Feb 2019	-3, -16	144, -72	16, 16	12, 13	O
Manam, Sabancaya, Mexico, Chile	19 Mar 2019	-3,-16,18,-24	144,-72,-98,-68	17, 16, 18, 15	9, 12, 6, 7	O
Sabancaya, Manam, Nev.Ruiz, Gamalama	20 Apr 2019	-16, -3, 5, 1	-72, 144,-75,128	17, 16, 16, 16	31, 12, 15, 7	O, T
Sinabung, Manam, Sabancaya	25 May 2019	3, -3, -16	98, 144, -72	17, 16, 16	11, 20, 21	O, T
Raikoke	22 Jun 2019	48	153	17	196	O
Raikoke, Ulawun	29 Jun 2019	48, -5	153, 151	15, 19	221, 107	O, T
Ubinas, Raikoke, Manam	19 Jul 2019	-16, 48, -3	-71, 153, 144	15, 16, 17	72, 141, 15	O, T
Ulawun, Mexico	3 Aug 2019	-5, 20	151, -100	19, 17	111, 12	O
Ubinas	16 Aug 2019	-16	-71	16	27	O, T

Table 2: Inventory of volcanic SO$_2$ emissions into the stratosphere integrated above 14 km in low latitudes, 13 km in mid-latitudes and 12 km in high latitudes. Listed altitudes and latitudes represent the region of maximum injection (or perturbation). Derived from satellite data (2002–2012) by MIPAS (M) and GOMOS (G). Based on a previous study from Brühl et al. (2018) with scaling factors for T63 and already published in an earlier version in Bingen et al. (2017) *(in italics)*. Extended with satellite data from SAGE II(V7.00) (S) back to 1990–2002, and from 2012–2019 by OSIRIS (O). Sometimes also TOMS/OMI/OMPS (T) are used for handling data gaps. For detailed description see the text. Data available online in Fortran formatted form and as netcdf (https://doi.org/10.26050/WDCC/SSIRC_3).

5 Implementation of SO$_2$ ~~from the Volcanic Sulfur Emission Inventory~~ emissions into the EMAC Model

The amount of SO$_2$ injected from each volcanic eruption is calculated by integrating the vertical SO$_2$ profiles, described in Table 2. Then, the SO$_2$ plumes are incorporated into the model simulations by adding the satellite-derived 3-dimensional perturbations of SO$_2$ mixing ratios to the simulated SO$_2$ at the time of the eruptions. In order to get the correct altitude distribution and to reduce additional errors caused by the low temporal resolution of the satellite data and possible numerical problems due to huge gradients or values out the range of used procedures in the model, we did not implement the volcanic SO$_2$ emissions as point sources. ~~In addition, effusive~~

Effusive eruptions and quiescent degassing volcanoes from the time-dependent monthly 3D climatology of Diehl et al. (2012) were added to the tropospheric SO$_2$ background emissions in the model simulations and truncated at an altitude of 200 hPa to avoid double counting in the stratosphere and uppermost troposphere ~~(Brühl et al., 2018).~~ since the original climatology also contains contributions of explosive volcanoes listed in Table 2 (only 1990 to 2009) in a crude way (Brühl et al., 2018). In some

cases, especially in the tropics, some SO$_2$ from degassing is transported by convection to the lowermost stratosphere (see e.g.
370 1998 in Figure 6).

The SO$_2$ emissions of the volcanic sulfur emission inventory are used in the EMAC model simulations, resulting in the time series shown in Figure 6, with mixing ratios between background conditions of a minimum of 0.001 ppbv (parts per billion by volume (10^{-9})) in volcanically quiescent periods, and highly active volcanic conditions with a maximum of 114 ppbv (as indicated at the top of the colour key, 5-day average) after the Pinatubo eruption. In the Junge-aerosol layer (i.e. around 25 to
375 29 km), typical mixing ratios of SO$_2$ are about –

0.03 ppbv.

EMAC Simulations of SO$_2$ in ppbv at 17 km 1991−2019

EMAC Simulations of SO$_2$ in ppbv at 20° S−20° N 1991−2019
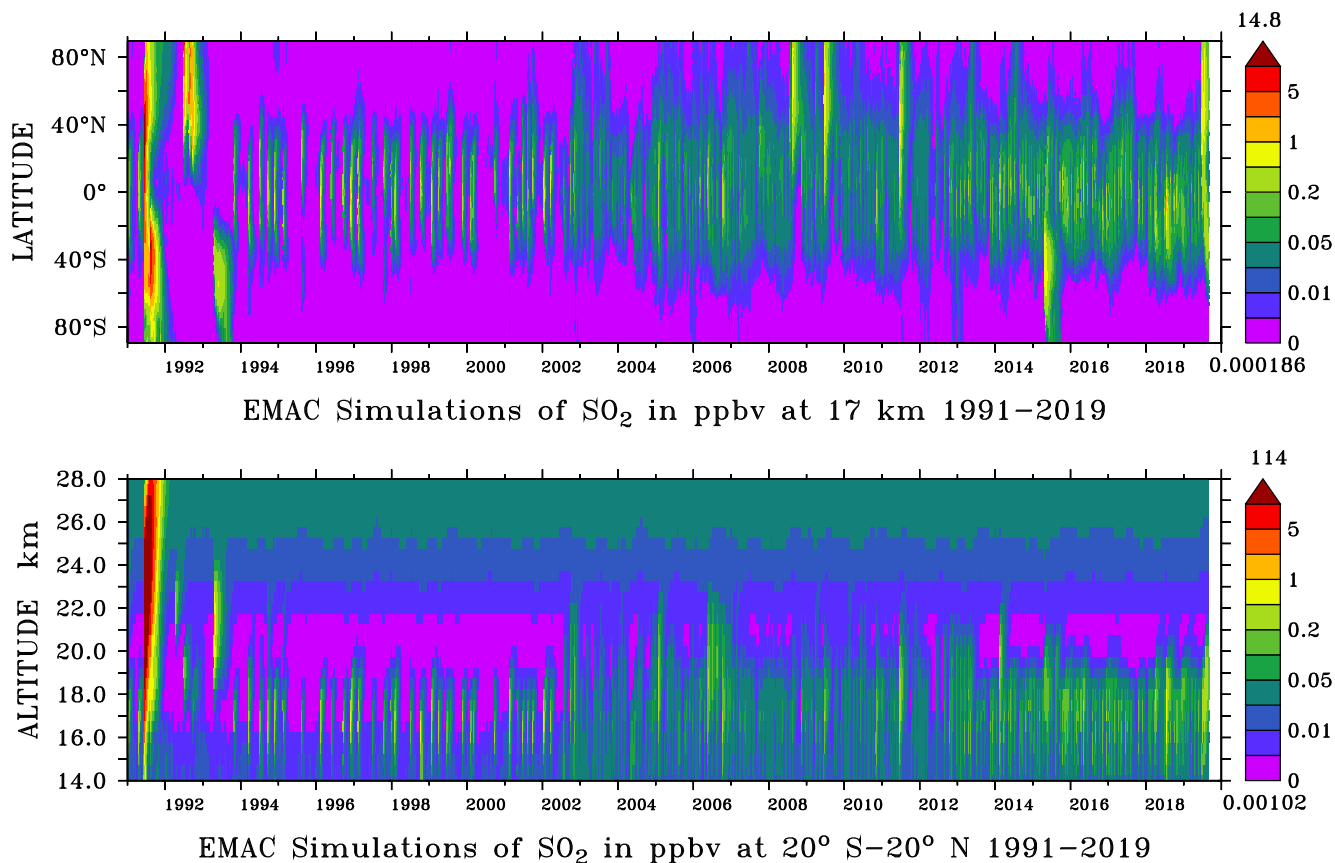
Figure 6. EMAC simulation of the stratospheric SO$_2$ mixing ratios (ppbv) (January 1991–August 2019) from the volcanic sulfur emission inventory (Table 2), in horizontal T63 resolution at 17 km altitude (top) and in vertical distribution for tropical regions 20° S–20° N (bottom). Maximum and minimum values are indicated above (dark red) and below (violet) the colour keys, respectively.

The volcanic eruptions in 1990 are entered included into the model during the spin-up phase of the model simulations (not shown), with the emissions of the first entry in Table 2 set to the upper limit –consistent to the Smithsonian reports, SAGE and TOMS. The low number of volcanic eruptions in 1991 and the following years is due to the low coverage of satellite data and

380   "saturation" effects of the satellite instrument (See subsection 2.4 about SAGE). The signatures of medium and small volcanic eruptions are too weak to be seen during the high concentrations in the first years after the Pinatubo eruption. There are also less entries in the Smithsonian database. From 2002 onwards, a higher number of small volcanic eruptions is captured in the volcanic sulfur emission inventory. This is rather due to the improved data coverage enabled by a larger number of satellite instruments, than to higher volcanic activity.

385   In most cases, the stratospheric SO_2 mixing ratios are highest at tropical latitudes. For this reason, tropical regions (20° S– 20° N) are chosen for the vertical distribution in the lower illustration of Figure 6 and subsequent figures. Exceptions to this typical SO_2 pattern are single medium strong volcanic eruptions at high latitudes like Kasatochi (2008), Sarychev (2009) and Raikoke (2019) in the northern hemisphere or Calbuco (2015) in the southern hemisphere. Another noteworthy case is the Nabro (2011) eruption, where the volcanic emissions were transported from the tropics to northern latitudes by the Asian

390   monsoon circulation .(Clarisse et al., 2014).

    MIPAS typically captures background SO_2 mixing ratios in the lowermost tropical stratosphere at 16 km to 17 km of around to 0.02 ppbv to 0.05 ppbv (Figure 1), which can be reproduced by the model only if many more volcanoes are considered than listed in the National Aeronautics and Space Administration (NASA) SO_2 database (Brühl et al., 2015). Some time periods with low volcanic activity resulting in stratospheric background conditions can be identified between 1996 and 2004. To reach

395   realistic SO_2 mixing ratios in the lower tropical stratosphere during these years, the oxidation of DMS and other sulfur species is needed. The lower panel of Figure 6 shows increasing SO_2 with altitudes of above 23 km due to additional production from OCS photolysis.

    The comparison of the simulated and observed SO_2 values shows that the volcanic SO_2 emissions from the volcanic sulfur emission inventory in Table 2 correlate well with the peaks of the mixing ratios in Figure 6, as they dominate the stratospheric

400   sulfur burden. In the stratosphere, SO_2 is converted to sulfate aerosol, which explains most of the interannual variability of the stratospheric aerosol burden as well as its influence on the stratospheric radiation. Generally, the conversion from of SO_2 to sulfate aerosol particles takes about two weeks in the UTLS and about two months in the lower and middle stratosphere. depends on several factors, such as the altitude, latitude, or season of the eruption and takes according to Höpfner et al. (2015) about 13, 23 and 32 days in 10–14, 14–18 and 18–22 km altitude, respectively, in midlatitudes. Carn et al. (2016) report an e-folding

405   time varying between 2–40 days. The range agrees with our simulations (and assumptions in section 4). Enhanced SO_2 concentrations from Pinatubo via photolysis of gaseous H_2SO_4 remained in the mesosphere for several years (Brühl et al., 2015) (Brühl et al., 2015; Rinsland et al., 1995).

6   Climate impact of stratospheric aerosol in EMAC model simulations

We compared the global influence of sulfur emissions on different atmospheric optical parameters. Based on Mie-theory-

410   lookup tables, optical properties such as optical depths, single scattering albedos and asymmetry factors, which are used in radiative transfer simulations, were calculated online for different aerosol types: sulfate, dust, organic carbon and black carbon, sea salt, and aerosol water .(Dietmüller et al., 2016). Via multiple calls of the radiation module RAD with and without aerosol
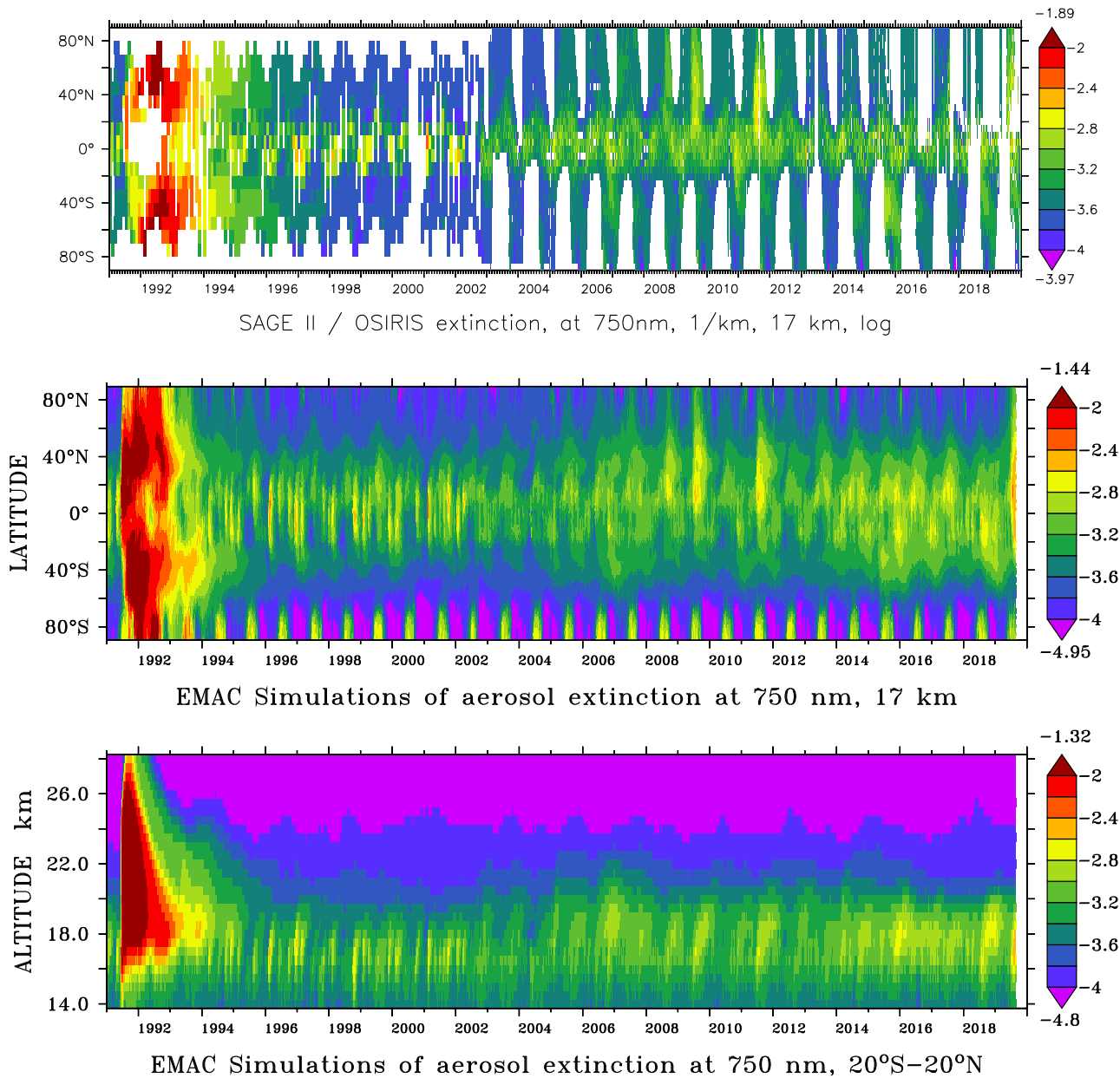
Figure 7. Comparison of aerosol extinction for 750 nm wavelength at 17 km altitude between the model simulations (lower panels) and SAGE II and OSIRIS satellite data (upper panel). EMAC simulation of the stratospheric aerosol extinction in logarithmic scale log(1/km) for 750 nm wavelength from January 1991–August 2019 based on the volcanic sulfur emission inventory (Table 2), in horizontal T63 resolution of zonal mean at 17 km altitude (middle) and in vertical distribution for tropical regions 20° S–20° N (bottom). Maximum and minimum values appear above (dark red) and below (violet) the colour keys, respectively.

the influence of stratospheric aerosol on instantaneous radiative forcing and heating is computed online (section 3). Also, the feedback to atmospheric dynamics is included.

6.1   EMAC ~~model~~ simulations of the stratospheric aerosol extinction

~~and~~ Figure 7 and Figure 8 show the global stratospheric aerosol extinction coefficients (in decadal logarithm) for the period 1991—2019 at ~~and~~ 750 nm and 550 nm wavelengths of the EMAC model simulations at 17 km altitude and its vertical profile for tropical regions for 20° S–20° N. For medium eruptions, the maximum of the aerosol extinction lies at an altitude between 16 km and 18 km. For this reason, an altitude of 17 km is chosen in the following analyses.

~~EMAC simulation of the stratospheric aerosol extinction in logarithmic scale log(1/km) for wavelength from January 1991–August 2019 based on the volcanic sulfur emission inventory (), in horizontal T63 resolution of zonal mean at altitude (top) and in vertical distribution for tropical regions 20° S–20° N (bottom). Maximum and minimum values appear above (dark red) and below (violet) the colour keys, respectively.~~

~~The EMAC model simulations of the aerosol extinction coefficients at~~ Figure 7 contains also the extinctions observed by OSIRIS and SAGE (~~) agree well with the satellite measurements of GOMOS () and SAGE II () for the aerosol layer at an altitude of where measured extinction values exceed ≈~~ interpolated from the observations at 550 and 1025 nm). The strongest event in these model simulations is the Pinatubo (Table 2) eruption in 1991, which dominates the stratospheric aerosol extinction coefficient for more than three years after the eruption with a global distribution from the equator to the poles in both hemispheres and a maximum altitude of more than 26 km. All other eruptions are significantly smaller, and for this reason a logarithmic scale is chosen. For further comparisons at 750 nm with GOMOS and OSIRIS we refer to Figure 3, Figure 4 and Brühl et al. (2018).

~~The~~

The EMAC model simulations of the aerosol extinction ~~coefficient at wavelength are required for the comparison with GOMOS~~ coefficients at 550 nm (~~) and OSIRIS () .~~ Figure 8) agree well with the satellite measurements of GOMOS (Figure 2) and SAGE II (Figure 5) for the aerosol layer at an altitude of 16–22 km where measured extinction values exceed ≈ 2×10^{-4} km^{-1}. Above about 24 km EMAC is lower than the observations because in the model meteoric dust particles were not considered.

The two figures show a similar distribution of the aerosol extinction at wavelengths of 550 nm and 750 nm. Due to the typical size and composition of stratospheric aerosol particles, the aerosol extinction is higher at 550 nm than at 750 nm. The peaks caused by mineral dust particles during summer in the Northern subtropics are more pronounced at 750 nm than at 550 nm.

~~EMAC simulation of the stratospheric aerosol extinction in logarithmic scale log(1/km) for wavelength from January 1991–August 2019 based on the volcanic sulfur emission inventory (), in horizontal T63 resolution of zonal mean at altitude (top) and in vertical distribution for tropical regions 20° S–20° N (bottom). Maximum and minimum values appear above (dark red) and below (violet) the colour keys, respectively.~~

Despite the presence of volcanoes in the Antarctic (like Mount Erebus), the seasonal change of extinction coefficient is not due to volcanic eruptions, but to the presence of Polar Stratospheric Clouds (PSCs) as simulated by the model.
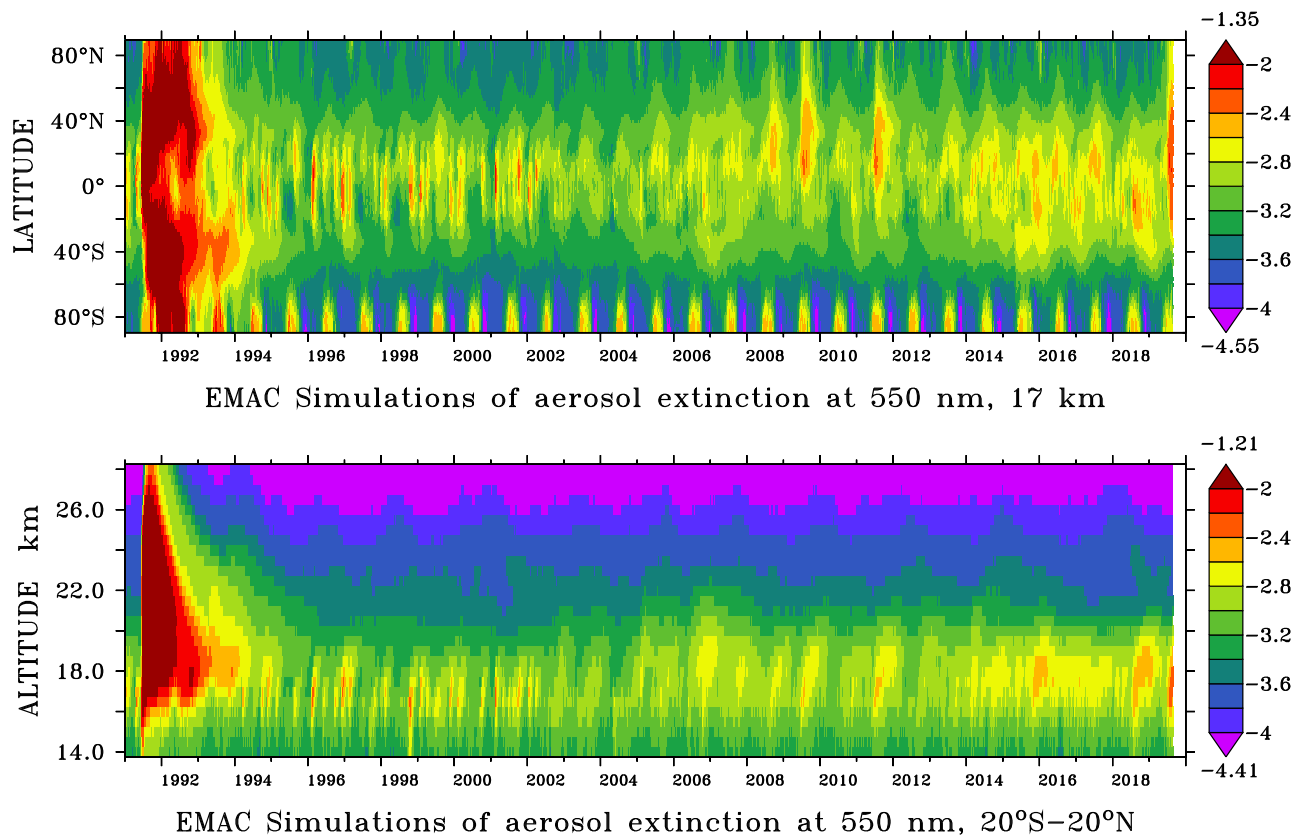
Figure 8. EMAC simulation of the stratospheric aerosol extinction in logarithmic scale log(1/km) for 550 nm wavelength from January 1991–August 2019 based on the volcanic sulfur emission inventory (Table 2), in horizontal T63 resolution of zonal mean at 17 km altitude (top) and in vertical distribution for tropical regions 20° S–20° N (bottom). Maximum and minimum values appear above (dark red) and below (violet) the colour keys, respectively.

6.2 EMAC ~~model~~ simulations ~~comparing the~~ of Aerosol Optical Depth (AOD)

The total stratospheric Aerosol Optical Depth (AOD) is obtained by the vertical integral of the aerosol extinction above an altitude of about 16 km (for mid-latitudes above about 14 km). The stratospheric AOD is shown on a logarithmic scale in Figure 9 and Figure 10 with the new model simulations (red line) compared to satellite observations (light blue, green, and blue lines). ~~Unlike most other studies, the stratospheric AOD is compared at the original wavelengths derived from different optical channels of the satellite instrument measurements. This~~ Using the wavelengths of the satellites in the calculations (Section 3) avoids introducing additional errors through the use of conversion factors to adjust the values between the different wavelengths.

From 1991 to 2012, SAGE II (light blue line), GOMOS (green line) and SAGE+CALIPSO and SAGE+OSIRIS (blue line) provide satellite data at a wavelength of 550 nm (Figure 9, OSIRIS data were converted from 750 nm by Glantz et al. (2014)).
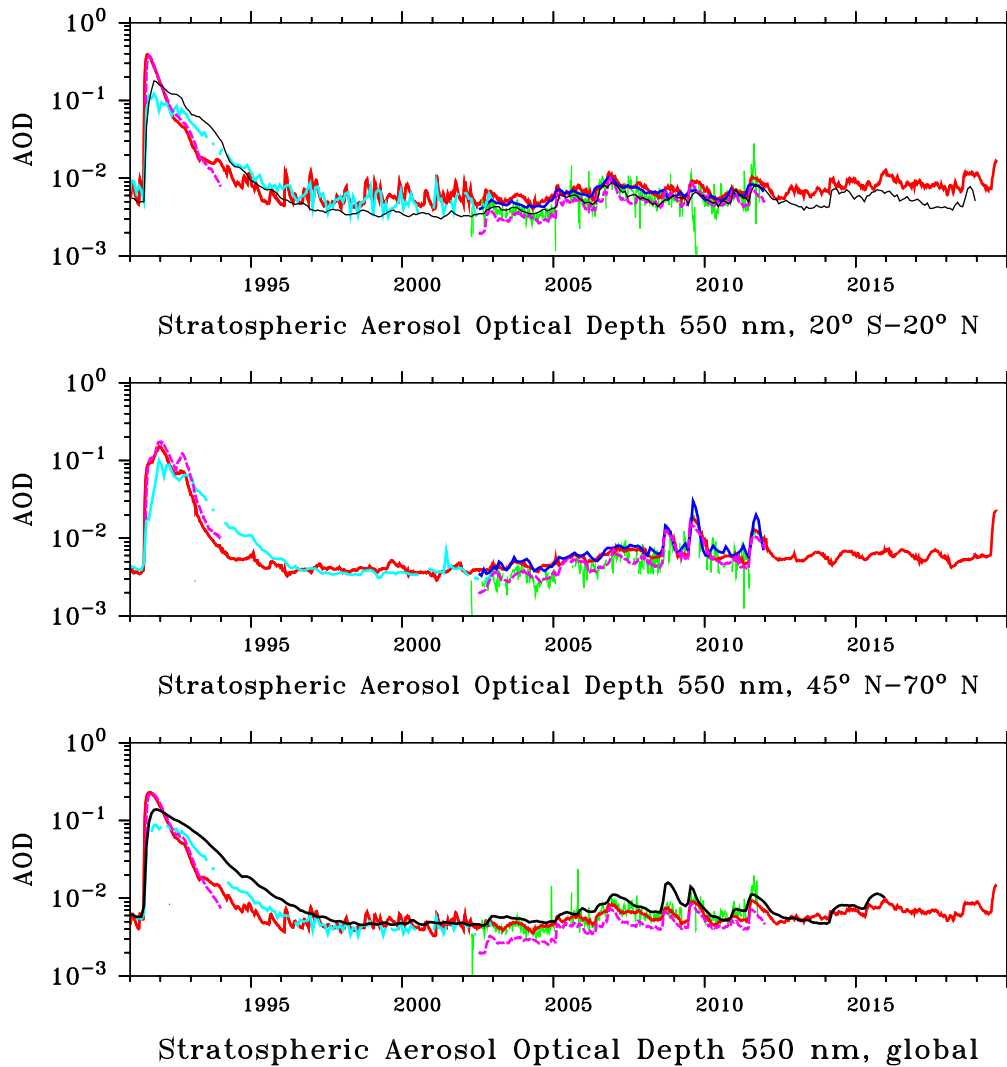
Figure 9. ~~The stratospheric Aerosol Optical Depth~~ Stratospheric AOD at 550 nm wavelength~~for tropical~~ : Tropical regions 20° S–20° N above 110 hPa ~~is~~ are shown on the top~~and for~~, the northern hemisphere 45° N–70° N above 145 hPa is shown in the middle and global means at the bottom. Satellite observations from SAGE II (Thomason et al., 2008) are indicated by the light blue line, GOMOS (Bingen et al., 2017) by the green line and values derived from SAGE+CALIPSO (upper figure) (Santer et al., 2014) and SAGE+OSIRIS (~~lower~~ middle figure) (Glantz et al., 2014) by the blue line. The red line shows the EMAC model simulations using the SO_2 injections of Table 2, compared to the simulations of Brühl et al. (2015) (pink dashed line) and the global stratospheric AOD from Schmidt et al. (2018) (black line in lower panel). The black line in the upper panel is from GloSSAC.

For convenience also GloSSAC (Kovilakam et al., 2020) is included as black line in the upper panel. Note the odd downward jump after 2012 which is not visible in OSIRIS data.

The maximum is reached after the Pinatubo eruption with a stratospheric AOD of 0.4 in the tropics (Figure 9 upper panel, EMAC), being an order of magnitude larger than the following medium eruptions with a stratospheric AOD of about 0.01 (e.g. Manam in early 1997, Rabaul in 2006 and Nabro in 2011). The differences after the large Pinatubo eruption in 1991 between the model simulations and the SAGE II observations are related to the "saturation" effects of the satellite instrument (i.e. data gaps due to an opaque path through the atmosphere at the tangent point) and can be observed for more than one year, (also shown above in Figure 5). In this study about 17 Tg SO$_2$ are injected for the Pinatubo eruption (Guo et al., 2004). Model comparisons by Timmreck et al. (2018) show that the span of used injections varies between 10 Tg SO$_2$ (e. g. Dhomse et al. (2014); Mills et al. (2016); Schmidt et al. (2018)) and 20 Tg SO$_2$ (e.g. English et al. (2013)). Thus, this study is in the middle range of the injected sulfur mass. On the other hand, filling the gaps in the SAGE data just by horizontal linear interpolation increases the peak AOD by about a factor of 2, which is close to the GloSSAC compilation. In Figure 10 the AVHRR (Advanced Very High Resolution Radiometer) by Long and Stowe (1994) at 630 nm are included, which are close to our simulations (if converted to 750 nm their AOD would be slightly less or to 550 nm slightly larger). When comparing the EMAC simulations (red line in Figure 9) with the simulation of Schmidt et al. (2018) (black line in Figure 9, lower panel) it can be recognized that a smaller value for the peak of the Pinatubo eruption occurs, but here it needs to be considered that Schmidt et al. (2018) are using monthly global-means. This has the consequence that the signal of single eruptions is blurred and smaller sized eruptions cannot be easily identified.

Between 1993 and 1996 the reduction of the stratospheric AOD in the model simulations is faster than ~~in~~ indicated by the satellite observations and in Schmidt et al. (2018). This indicates that the removal of stratospheric aerosol is still too ~~fast~~ rapid from applying the modal model. Schmidt et al. (2018) show a slower decrease in AOD after the Pinatubo eruption. This could indicate that EMAC still needs better fine-tuning of the size distribution modes, or adding modes in the aerosol submodel to improve the aerosol removal in the stratosphere. Here the sectional aerosol model used by Schmidt et al. (2018) might have an advantage (Clyne et al., 2021). Additionally, smaller volcanic eruptions might be missing, in view of the low number of identified events in the years after the Pinatubo eruption.

In Figure 10, the coverage of GOMOS (green line) is often too low at a wavelength of 750 nm for the years from 2002 to 2012, so the inclusion of OSIRIS data (blue line) is important (Brühl et al., 2018). For the years after 2012 the timeline only contains data from OSIRIS at 750 nm wavelength.

Nevertheless, there still remain small differences between the model simulation and the observations, for instance in 2010, which indicates missing volcanic eruptions (or an underestimation of the Merapi eruption by MIPAS compared to OSIRIS, Appendix B).

The different distributions of the peaks in the upper and the lower ~~figures~~ panels is related to the latitude of the volcanic eruptions. Emissions reaching the stratosphere from strong eruptions in the tropics are distributed by the Brewer-Dobson circulation over the northern and southern hemisphere even to high latitudes, as in the cases of Soufriere Hills and Rabaul in 2006. However, if an eruption takes place at high latitudes (such as for Redoubt 2009) or at mid-latitudes like Kasatochi (2008)~~or~~, Sarychev (2009) or Raikoke (2019), most of the emissions stay in the northern hemisphere and the signal in the tropics
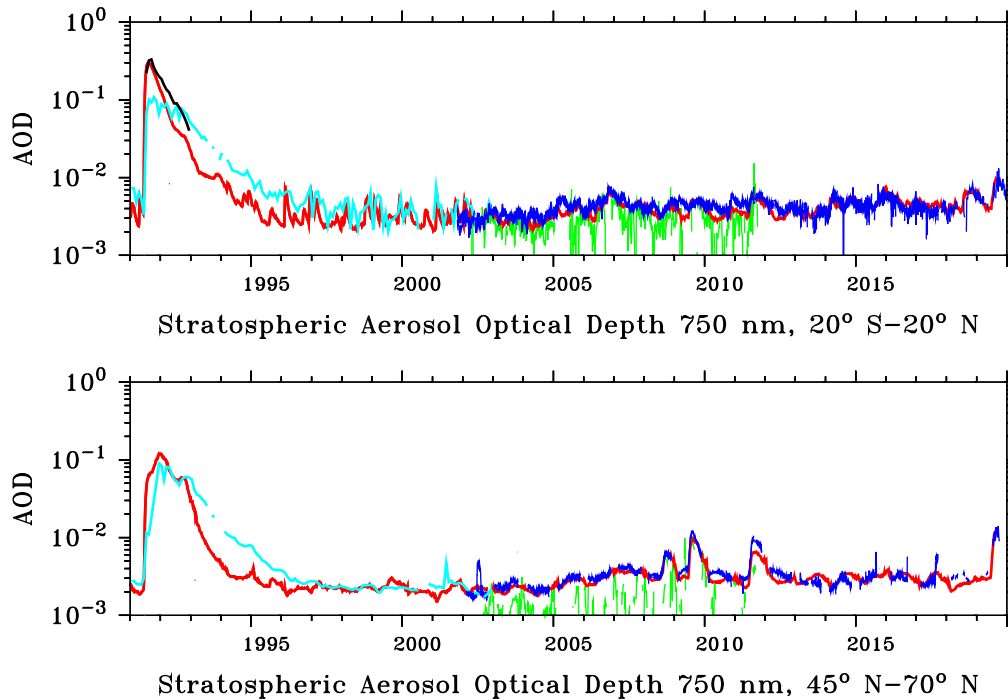
Figure 10. ~~The stratospheric Aerosol Optical Depth~~ Stratospheric AOD at 750 nm wavelength~~for tropical~~: Tropical regions 20° S–20° N above 110 hPa ~~is~~ are shown on the top and for the northern hemisphere 45° N–70° N above 145 hPa at the bottom. Satellite observations from OSIRIS (Rieger et al., 2019) are indicated by the blue line and GOMOS (Bingen et al., 2017) by the green line. For the Pinatubo period the black line shows the AVHRR observations by Long and Stowe (1994) at 630 nm (upper panel). The light blue line shows the interpolation of SAGE data at 550 nm and 1025 nm wavelengths. The EMAC model simulations, using the SO_2 injections of Table 2, are shown by the red line.

is weaker. Our northern hemisphere results for AOD of about 0.025 for Raikoke (550nm) agree within uncertainties with Kloss et al. (2021) who use different satellites and different modelling approaches.

6.3 EMAC ~~model~~ simulations comparing the radiative forcing at the tropopause

495 The instantaneous radiative forcing of the stratospheric aerosol is calculated by multiple calls of the RAD submodel (section 3). The simulated global negative radiative forcing in Wm^{-2} of stratospheric aerosol at the tropopause is illustrated in Figure 11.

As the Pinatubo eruption caused a negative radiative forcing of more than ~~a~~ an order of magnitude greater than all other eruptions since then, the y-axis of the lower figure is plotted in inverted logarithmic scale; for instance, 10^0 corresponds to a global negative radiative forcing of $1.0\,Wm^{-2}$. The new model simulations with the additional volcanic eruptions (red line)

500 are closer to the calculated estimates from satellite extinction measurements of SAGE, GOMOS and CALIOP ~~(Cloud-Aerosol Lidar with Orthogonal Polarization)~~ by Solomon et al. (2011) (green ~~crosses~~bars) than in previous studies (e.g., Brühl et al.
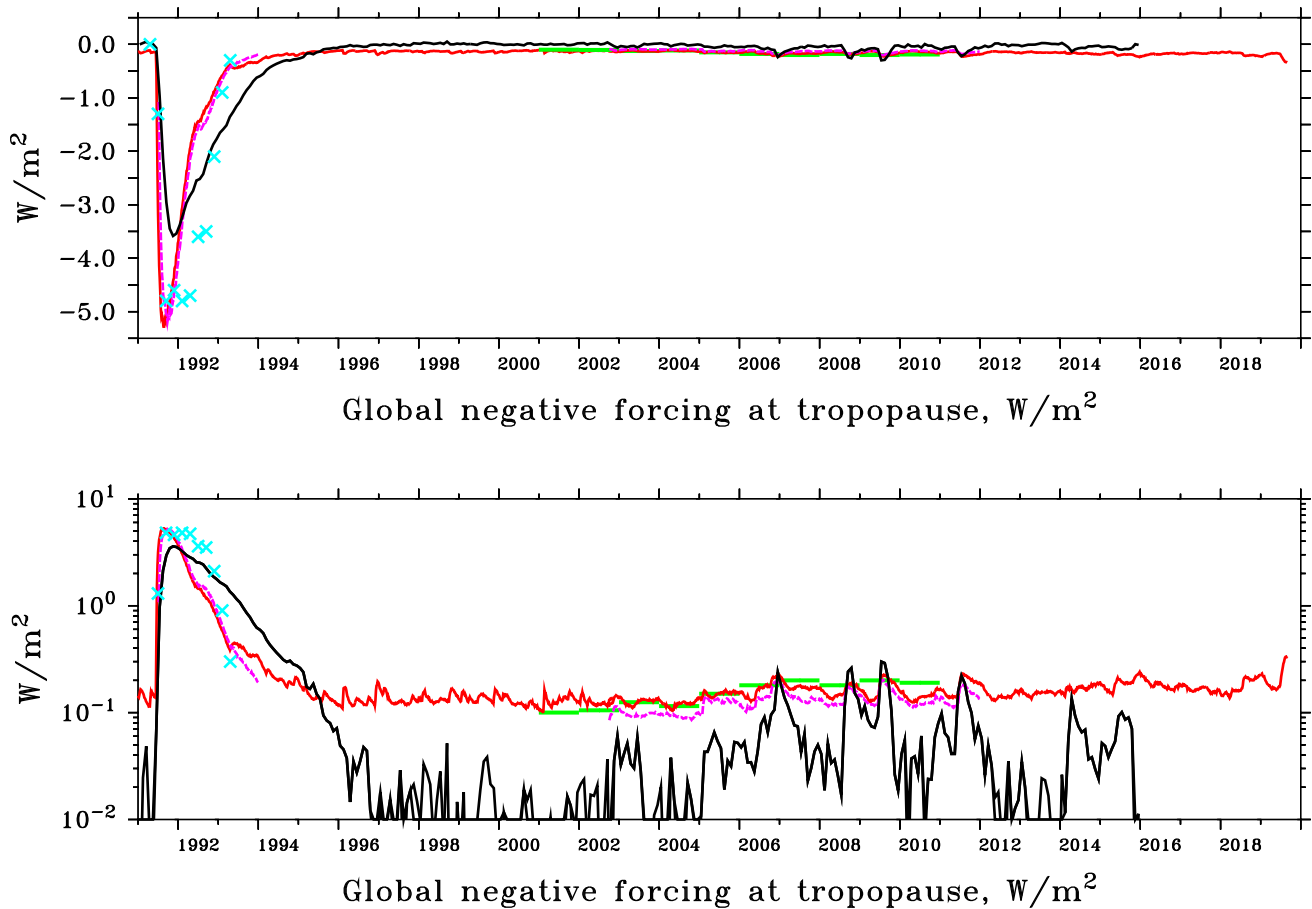
Figure 11. Global negative radiative forcing ~~at the tropopause (, solar + IR)~~ by stratospheric aerosol. Estimated averages for solar forcing at the top of the atmosphere from satellite observations of the Earth Radiation Budget Experiment (ERBE) ~~(Wong et al., 2006)~~ (72-day means) (Wong et al., 2006; Toohey et al., 2011) are indicated by light blue crosses and annual averages derived from observations by Solomon et al. (2011) as green ~~crosses~~bars. The EMAC model simulations with instantaneous forcing at the tropopause (185 hPa, solar + IR) based on volcanic SO_2 emissions are represented by the red line, compared to the simulations of Brühl et al. (2015) (pink line) and data from Schmidt et al. (2018) with volcanic effective radiative forcing (black line).

(2015) ~~). This~~ pink line). A comparison with data of total volcanic effective radiative forcing from (Schmidt et al., 2018) is shown by the black line. The large difference to our simulation in periods between medium sized eruptions shows that even medium- and small-sized volcanic eruptions can reach the stratosphere and have a significant influence on the global radiative forcing at the tropopause. In the period considered here, the volcanoes are the dominant factor in instantaneous global negative radiative forcing, with up to $0.2\,\mathrm{Wm^{-2}}$ for Rabaul and Nabro (2011) and more than $0.3\,\mathrm{Wm^{-2}}$ for Raikoke/Ulawun (2019) compared to the volcanically quiescent period in 2002. ~~Nevertheless, there remain small differences between the model simulation and the observations, for instance in 2010, which indicates missing volcanic eruptions (or an underestimation of the~~

~~Merapi eruption by MIPAS). The strongest~~ The value for Raikoke/Ulawun is within the range discussed in Kloss et al. (2021)
. The strongest instantaneous global radiative forcing in the model simulations is caused by the Pinatubo eruption with a maximum of about -5 Wm^{-2}; this is in good agreement with the results of Minnis et al. (1993) ~~. However, the same effect can be seen~~ and the observations of the ERBE satellite (light blue crosses in Figure 11~~as in ; that is,~~). A comparison between the ERBE observations (light blue crosses) and Schmidt et al. (2018) shows that they may underestimate the maximum of the ~~global negative radiative forcing drops again too quickly after the Pinatubo eruption . For the Pinatubo case, the particle fraction in the coarse mode is probably overestimated, resulting in an overestimation of sedimentation by large particles, even though the boundary between the accumulation and coarse modes in the aerosol module GMXe was shifted from a diameter of to to represent stratospheric conditions with long residence times (Brühl et al., 2015)~~Pinatubo eruption 1991 but one has to keep in mind that ERBE sees only the solar forcing which is larger than total forcing. Schmidt et al. (2018) estimates the forcing from the difference of a simulation with and without Pinatubo, i.e. including dynamical and chemical adjustments which leads to a smaller value than instantaneous forcing at the peak. A similar approach with EMAC leads to a peak forcing of about -4 Wm^{-2} (but with nudging of the troposphere in contrast to Schmidt et al. (2018) which leads to higher forcing here).

6.4 EMAC ~~model~~ simulations of the stratospheric aerosol radiative heating

The simulated instantaneous aerosol radiative heating in the model is derived from multiple radiation calls with and without aerosol in the radiation submodel RAD. Aerosol formation is calculated by the GMXe submodel, while the aerosol optical properties are calculated by the AEROPT submodel.

Figure 12 shows the calculated local heating effects in the stratospheric aerosol layer. Small and medium volcanic eruptions have the largest effects between altitudes of 17 km and 18 km and generate atmospheric heating of up to 0.03 K/day. The eruption of Pinatubo, on the other hand, had significantly stronger effects at altitudes of 20 km to 25 km and caused atmospheric heating of more than 0.7 K/day, which corresponds quite well with the results of Rieger et al. (2020) showing a maximum of instantaneous solar heating rate of 0.5 K/day in the tropics near 24 km plus thermal heating rates of about 0.2–0.3 K/day. This is about 23 times greater than all other eruptions in the model simulation, including the Raikoke eruption in 2019. ~~After such a strong volcanic eruption, the enhanced radiative heating exerts an effect on dynamics, influences the global spread of the volcanic cloud and leads to an upward transport of the aerosol itself as well as of other chemical tracers including ozone (Timmreck et al., 1999).~~

Further, a seasonal signal contributes significantly to the radiative heating in the northern subtropics. This is caused by transport of desert dust to the UTLS mostly via the Asian summer monsoon convection, which generates additional heating during the time of the Asian summer monsoon (Brühl et al., 2018).
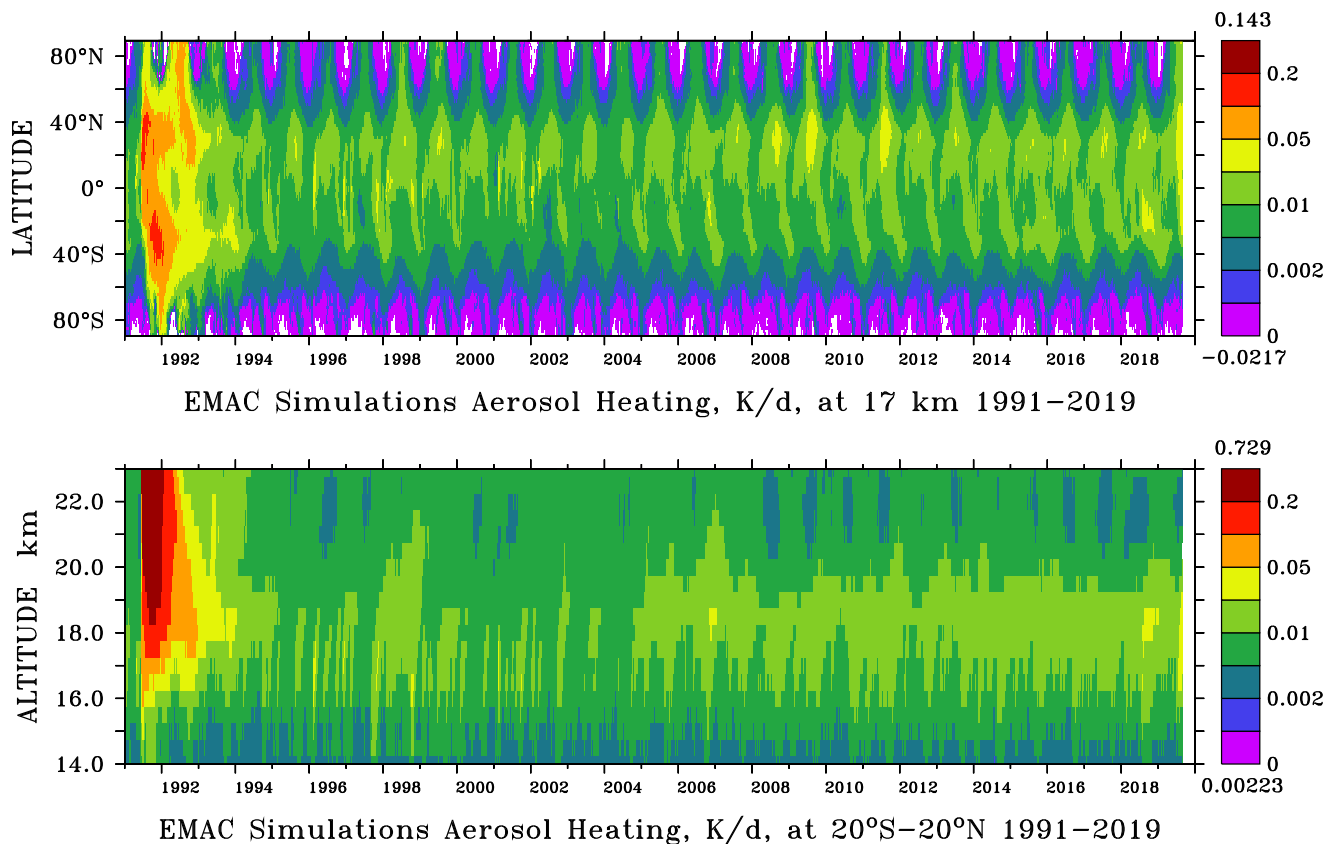
Figure 12. EMAC simulation of the aerosol radiative heating in K/day for solar and infrared radiation from January 1991–August 2019 based on the volcanic sulfur emission inventory (Table 2), in horizontal distribution at 17 km altitude (top) and in vertical distribution for tropical regions 20° S–20° N (bottom).

7 Discussion and conclusions

The objective of this study was to generate a detailed volcanic sulfur emission inventory and to improve the EMAC model
540 simulations of the global stratospheric aerosol and sulfate burden, and compute the volcano-induced radiative forcing through validation with satellite data.

~~Compared to previous studies by Brühl et al. (2015) and Bingen et al. (2017), the horizontal resolution in these new model simulations is increased from T42L90 to T63L90. Since the convection parametrisation of the model was developed for the higher resolution T63L90, the stratospheric aerosol distribution is also in much closer agreement with the satellite observations~~
545 ~~() than before (Brühl et al., 2018).~~

~~Due to the newly available satellite data sets of SAGE II and OSIRIS, the time series could be extended in both directions to~~ Updated OSIRIS data allowed us to extend the comparisons of Brühl et al. (2018) to 2019; a detailed analysis of L2 SAGE II data together with the Smithsonian volcano database was used to extend the comparison back to 1990. So the simulations in

this paper encompass a total of 29 years and now covers the years 1991 to 2019, instead of 2002 to 2012 ~~in~~ as in our previous

550 work. Additional updated three-dimensional data sets are now available, which provide better temporal resolution than the commonly used monthly means of the MIPAS, GOMOS and OSIRIS instruments. The temporal resolution is now five days for the three instruments and it is possible to identify multiple volcanic eruptions within a short period of time. With the three-dimensional data sets, the vertical distribution of SO_2 can be distinguished and the amount of sulfur reaching the stratosphere can be calculated much more accurately than by estimation of a total column. To exclude tropospheric emissions and signals

555 from high-altitude clouds, the integration of the sulfur emissions is set above an altitude of 12 km at high latitudes, 13 km at mid-latitudes and 14 km at low latitudes. To avoid double counting, tropospheric sulfur emissions are treated separately.

Our volcanic sulfur emission inventory is an improvement on the version published by Bingen et al. (2017) and Brühl et al. (2018). While the previous version included 230 explosive volcanic eruptions, the new version now lists more than 500 eruptions. An overview of these eruptions is given in the improved volcanic sulfur emission inventory in Table 2, which also includes the

560 estimated stratospheric SO_2 emissions as well as the plume altitudes. These consist of about 80 eruptions in the first time period between 1990–2002 measured by the SAGE II instrument, 240 eruptions in 2002–2012 measured with multiple instruments and 230 eruptions in the last time period 2012–2019 measured by OSIRIS. ~~Older versions of the volcanic sulfur emission inventory did not incorporate enough data to allow assessment of the true volcanic contribution to the radiative forcing of the atmosphere and only spanned the period 2002–2012~~The inclusion of many more small size eruptions reaching the UTLS

565 has the consequence that stratospheric aerosol optical depth and radiative forcing does not decrease to almost zero between medium size eruptions, in agreement with observations, and in contrast to a previous work.

Strong volcanic eruptions can inject several teragrams of SO_2 directly into the stratosphere. For this reason, the maxima of the global stratospheric SO_2 concentrations correlate well with the eruption events of the volcanic sulfur emission inventory in Table 2.

570 The SO_2 emissions of smaller volcanic eruptions can reach the lower stratosphere by convective transport through the tropical tropopause, which results in accumulation of sulfate aerosol in the lower stratosphere. This was demonstrated to be ~~essential for correctly assessing the extinction coefficient~~ important for correctly modelling the AOD in volcanically quiescent periods.

Our analysis shows the importance of using multi-instrument satellite data sets to fill data gaps and to detect as many volcanic

575 eruptions as possible. The ~~best~~ optimal data coverage was in the time period from 2002 to 2012, for which simultaneous measurements from the MIPAS, GOMOS and OSIRIS instruments are available. ~~The~~ For 2002 to 2005 this includes also SAGE II. The periods with simultaneous observations by MIPAS and the other instruments were used to develop and validate a method for estimating injected SO_2 and its distribution from extinction observations. The evaluation by the satellite data sets shows that GOMOS is important for detecting volcanic eruptions in MIPAS data gaps and for a better attribution of individual

580 eruptions. Consequently, the combination of MIPAS, GOMOS and OSIRIS data leads to better SO_2 input ~~and~~ for calculating the radiative forcing in the chemistry climate model EMAC.

~~Our work also confirms the importance of improving the volcanic sulfur emissions inventory for the modelling of the radiative effects by stratospheric aerosols. The new model simulations demonstrate that volcanic emissions have a dominant~~

~~impact on the stratospheric aerosol burden and explain most of the interannual variability of the stratospheric aerosol extinction~~ ~~during the considered time period. These emissions have a large impact on the stratospheric aerosol forcing and heating, which~~ ~~influence the climate and the global circulation for a few years after big volcanic eruptions.~~

~~The aerosol radiative heating resulting from large volcanic eruptions like Pinatubo triggers enhanced tropical upwelling, which causes a lofting of the injected SO_2 and the aerosol as well as other chemical tracers. Here, the radiative feedback on dynamics is required for estimates of the aerosol extinction in the upper part of the volcanic aerosol plume that correspond with observations.~~

~~Big~~ Large volcanic ash plumes can interfere with the SO_2 signal in satellite measurements, and satellites could be "blind" during the first few days or months after an eruption (Höpfner et al., 2015). However, most volcanic ash particles are relatively large and sediment after some hours or days, so they have only minor climatic significance (Boucher, 2015) and are not discussed in detail here. There are, however, volcanoes (e.g. ~~Kelut,~~ eruption of Kelut in 2014 (Zhu et al., 2020)) which emit small ash particles which can stay in the lower stratosphere for several months (Vernier et al., 2016). Moreover, it has been found that the model setup might have to be improved by including an additional aerosol mode to slow down the removal by sedimentation of stratospheric aerosol after big volcanic eruptions.

Some satellite data sets still contain data gaps and noise. Comparing the model results with OSIRIS data in the northern tropics (Figure 10) indicates that some volcanic events are still underestimated or missing in the volcanic sulfur emission inventory in the year 2010. This could also explain the differences between the model simulations and satellite observations from Solomon et al. (2011), indicated by green ~~crosses~~ bars in Figure 11 of the radiative forcing in this year.

~~A study of ? shows that meteoric smoke is the dominant source of aerosol extinction in the upper stratosphere and mesosphere above in the tropics and above poleward of 30 degrees in latitude. The meteoric material could act as sink for gaseous H_2SO_4. Detected meteoric dust in the UTLS (?) also suggests that it has to be incorporated into the model to reproduce observations of higher aerosol concentrations in the upper stratosphere. Thus, the aerosol extinction for the upper stratosphere and mesosphere is underestimated in model simulations without implementation of meteoric material in the radiation calculations.~~

The model also includes mineral dust and organics from the troposphere transported up to the UTLS. The EMAC simulations show a seasonal signal in the stratospheric AOD and an enhanced radiative heating in the northern hemisphere, induced by the convective transport of mineral dust to the UTLS in the Asian monsoon region. This is confirmed by satellite observations and studies by Klingmüller et al. (2018). The influence of wildfires and other biomass burning plumes on stratospheric AOD has increased in recent years (Fromm et al., 2019), and this effect should be included in the model to account for perturbations in organic and black carbon.

~~In any case, the findings of the IPCC (2013) report are confirmed: frequent~~ Frequent volcanic eruptions of moderate and small intensities, injecting sulfur gases to the upper troposphere and lower stratosphere, contribute significantly to the stratospheric aerosol layer through accumulation. These cause a global negative radiative forcing of 0.12 (0.22 to 0.08) Wm^{-2} at the ~~surface, like~~ tropopause, for example in the case of the ~~volcanic~~ eruptions of Soufriere Hills/Rabaul (2006), Nabro (2011) and the combination of the Sinabung, Wolf and Calbuco eruptions (2015) with a radiative forcing of up to 0.13 Wm^{-2}, and 0.2 Wm^{-2} for Raikoke/Ulawun (2019) ~~. This shows that also medium-sized volcanic eruptions can contribute to a slowdown of~~

~~global warming like in the mid-2000s, thus improving the new model simulations, especially in 2007, a year for which several~~
620 ~~eruptions were not represented in previous studies.~~ compared to volcanically quiescent periods (e.g.in 2000 to 2002).

Code and data availability. The Modular Earth Submodel System (MESSy) is continuously developed and used by a consortium of institutions. The use of MESSy and access to the source code is licensed to all affiliates of institutions which are members of the MESSy Consortium. Institutions can become a member of the MESSy Consortium by signing the MESSy Memorandum of Understanding. More information can be found on the MESSy Consortium website (http://www.messy-interface.org,MessyConsortium,2017). The input data files
625 and model output of EMAC used here are stored at DKRZ, Hamburg, the volcanic inventory and the output for radiative forcing also at WDCC (https://doi.org/10.26050/WDCC/SSIRC_3).

For the MIPAS data we refer to http://www.imk-asf.kit.edu/english/308.php. The sulfur injections from the volcanoes were estimated utilizing the NASA SO_2 database at GSFC (http://so2.gsfc.nasa.gov) and the Smithsonian volcano database (http://www.volcano.si.edu).

Author contributions. JS, CBr and JL defined the scientific questions and scope of this work. JS and CBr conceived the idea and methodology
630 used in this paper and carried out the model simulations and the analysis. MH provided the MIPAS data, CBi provided the GOMOS data and LR provided the OSIRIS data. All authors participated in the scientific discussion in regard to satellite data in particular. JS wrote the manuscript, and all authors reviewed the manuscript and provided advice on the manuscript and figures.

Competing interests. The authors declare that they have no conflicts of interest.

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640 MIPAS, OSIRIS, SAGE, the Copernicus Climate Change Service (projects C3S_312a_Lot5 and C3S_312b_Lot2), and EOSDIS (NASA) teams for their productive cooperation and providing their satellite data sets. GloSSAC (V2) data were obtained from the NASA Langley Research Center - Atmospheric Sciences DataCenter. We are also grateful to our colleagues from the EMAC community and all other MESSy developers and users for their support. We thank A. Schmidt for editing our paper and providing the data of her paper for comparisons of our results. We would also like to thank the referees for their comments to help us improving our paper.

645 **Appendix A: List of MESSy submodels used in this study**

The computations for this study were performed on the Mistral supercomputer at the Deutsches Klimarechenzentrum (DKRZ), Hamburg, Germany. For this purpose, EMAC (ECHAM5/MESSy Atmospheric Chemistry), a coupled atmospheric circulation model consisting of the 5th generation of European Centre Hamburg general circulation model (ECHAM5) and the Modular Earth Submodel System (MESSy) was used.

650   ECHAM5 (5th generation of European Centre Hamburg general circulation model) is an atmospheric general circulation model (Roeckner et al., 2003, 2006), which runs with self-consistent quasi-biennial oscillation (QBO). It is nudged to the meteorological ERA-Interim reanalysis data of the European Centre for Medium-Range Weather Forecasts (ECMWF) up to about 100 hPa.

MESSy (Modular Earth Submodel System) is an earth system model, which consists of several submodels (Jöckel et al., 2005, 2006, 2010
655   . An overview of the submodels used in this study is given in Table A1.

Submodel	Function	Reference
AEROPT	Aerosol optical depth	Dietmüller et al. (2016)
AIRSEA	Air-sea exchange of trace gases	Pozzer et al. (2006)
CAABA/MECCA	Atmospheric chemistry	Sander et al. (2011)
CONVECT	Convection processes	Tost et al. (2006b)
CVTRANS	Convection transport of tracers	Tost et al. (2010)
DDEP	Dry deposition	Kerkweg et al. (2006a)
GMXe	Global Modal Aerosol eXtension	Pringle et al. (2010)
IMPORT	Import of external data files	Jöckel et al. (2006)
JVAL	Photolysis rate coefficients	Jöckel et al. (2006)
LNOX	NOx lighting production	Tost et al. (2007)
MSBM	Multiphase Stratospheric Box Model	Jöckel et al. (2010)
OFFEMIS	Off-line emissions	Kerkweg et al. (2006b)
ONEMIS	On-line emissions	Kerkweg et al. (2006b)
QBO	QBO nudging	Giorgetta et al. (2002)
RAD	RADiation	Dietmüller et al. (2016)
SCAV	Scavenging (wet removal)	Tost et al. (2006a)
SEDI	Aerosol sedimentation	Kerkweg et al. (2006a)
TNUDGE	Tracer nudging	Kerkweg et al. (2006b)
TROPOP	Tropopause calculation	Jöckel et al. (2006)

Table A1. List of used MESSy submodels. Reference and short description from http://www.messy-interface.org. Parts of the base model copied into MESSy which must always be active are not listed here.

Mineral dust emissions are calculated online using the emission scheme of Astitha et al. (2012) as part of the ONEMIS submodel. The convection was calculated with the CONVECT submodel (Tost et al., 2006b), with the convection scheme from

Tiedtke (1989) and the Nordeng (1994) closure. The convection parametrization is sensitive to the model resolution, which results in differences between different model resolutions in the vertical transport of tracers, like dust, water vapor, ozone and SO_2, especially near the low latitude tropopause (Brühl et al., 2018). To avoid a phase drift, we used the QBO submodel for weak nudging to the QBO zonal wind observations (Giorgetta et al., 2002). The loss of gas phase species to the aerosol is parametrized in the 3rd EQuilibrium Simplified Aerosol Model (EQSAM3) (Metzger and Lelieveld, 2007). The uptake of gases on wet particles and on acid aerosol particles is included in the model calculation. Concerning removal mechanisms, the SCAVenging submodel calculates the loss of atmospheric tracers and aerosols by wet deposition, as well as the liquid phase chemistry in clouds and precipitation (Tost et al., 2006a). The chemistry of the CAABA/MECCA submodel contains photolysis reactions, which need photolysis rate coefficients (J-values) for tropospheric and stratospheric species computed by the JVAL submodel. The RAD_FUBRAD sub-submodel is used to calculate the shortwave heating rates from the absorption of UV by O_2 and O_3 in the upper stratosphere and mesosphere.

Appendix B: Comparison of volcanic injections derived from simultaneous MIPAS, SAGE, GOMOS and OSIRIS observations

The eruption of Reventador in the tropics in November 2002 has shown to be an ideal case where simultaneous observations of all satellite sensors were available so that the direct SO_2 observation could be used for development and validation of a conversion formula for the 750 nm extinction seen by GOMOS and OSIRIS, which works also approximately for SAGE if its observations at 530 and 1025 nm are interpolated to 750 nm. Here we first use the ratio between model calculated sulfate volume mixing ratio and its share on extinction in low latitudes of the lower stratosphere which is typically $1.2\times10^{12}\times$ air density (in molecules/cm^3). This works for medium size eruptions and data available over about four weeks following the eruptions, and if no other events occur less than about four weeks before which is the case for the Reventador eruption. If the time lag of data is several weeks a correction factor >1 has to be applied to account for removal processes, if another event is relatively close in time, the factor has to be <1 to remove the influence of the previous event. For Reventador the factor is 1 (for OSIRIS 0.8 is slightly better). For all instruments the derived injected SO_2 mass is very close to 77 kt as shown in Table 2. The spatial patterns are similar, except when the zonal wind causes a shift in longitude due to the time lag from conversion of SO_2 to aerosol, see Figure B1. In the case of SAGE, the alternate method of Grainger et al. (1995) involving aerosol surface area density (SAD) and aerosol volume density is more suitable to remove cloud perturbation. It is assumed that sulfate mixing ratios correspond to the SO_2 injected. Some uncertainty remains from removing the background which we have done by subtracting a fraction of the derived SO_2 at the longitude where it has a minimum, i.e. the longitude where the effect of the volcano is smallest for all altitudes. Integrated injected SO_2 masses for all examples are provided in Table B1.

For the eruption of Merapi in November 2010 the satellite instruments do not agree. From OSIRIS about 70% more injected SO_2 is derived than from MIPAS, i.e. 170 kt instead of 97 kt used in the transient simulation (see Table 2 and differences in Figure 10). GOMOS has too sparse data here to obtain a proper integral directly but patterns are similar (Figure B2). If other
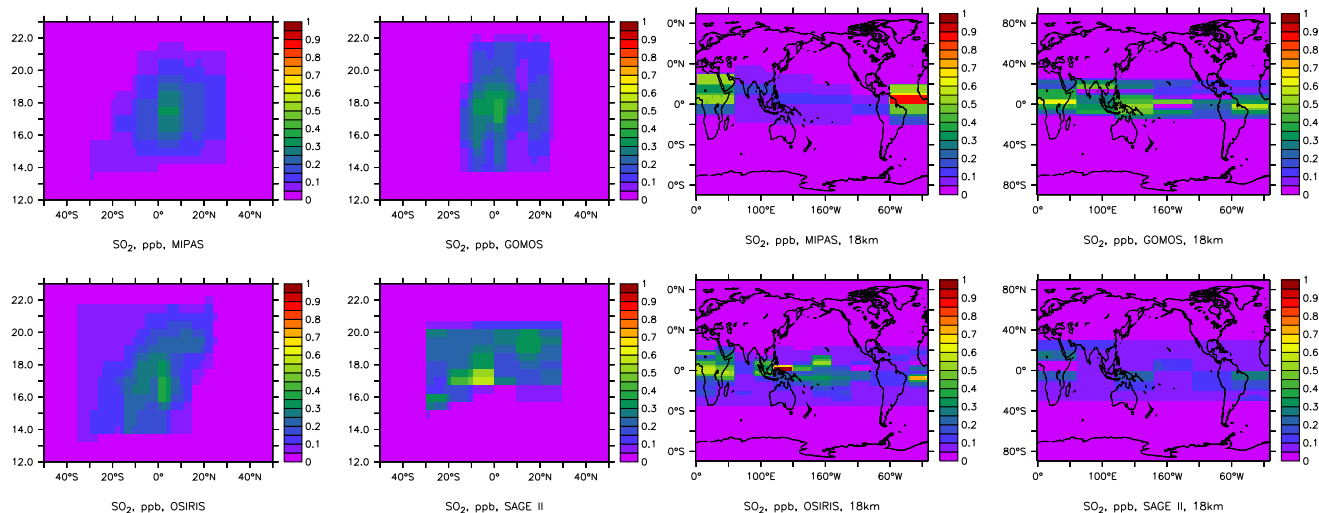
Figure B1. 2002 Reventador eruption: SO$_2$ derived from MIPAS and the other 3 instruments. Average vertical distribution (left) and plumes at 18 km (right). OSIRIS with correction factor 0.8 due to remnants from the Ruang eruption, SAGE II with the Grainger-based method.

Eruption	MIPAS	GOMOS	filled	OSIRIS	SAGE II
Reventador 2002	77	75	-	89	80
Merapi 2010	97	18	77	170	–
Sarychev 2009	446	141	-	353	–
Manda Hararo 2009	82	81	-	101	–

Table B1. Integrated mass of injected SO$_2$ in kt

690 information is available, the gaps can be filled with likely values in the region where the plume was seen, a method which had to be applied also to some events seen by OSIRIS in 2018 and 2019 for which the data were sparse.

For high latitude eruptions the longer conversion time of SO$_2$ to sulfate compared to the tropics has to be considered which, together with aerosol removal processes, lead to a weaker extinction signal. To account for this a correction factor of about two in the conversion formula for OSIRIS for example for Sarychev in June 2009 leads to values consistent to the ones derived by
695 MIPAS (Figure B3). For the low latitude eruption of Mando Hararo in the same entry of Table 2 (separated at 24° N for the integration) the factor 1 is still appropriate.
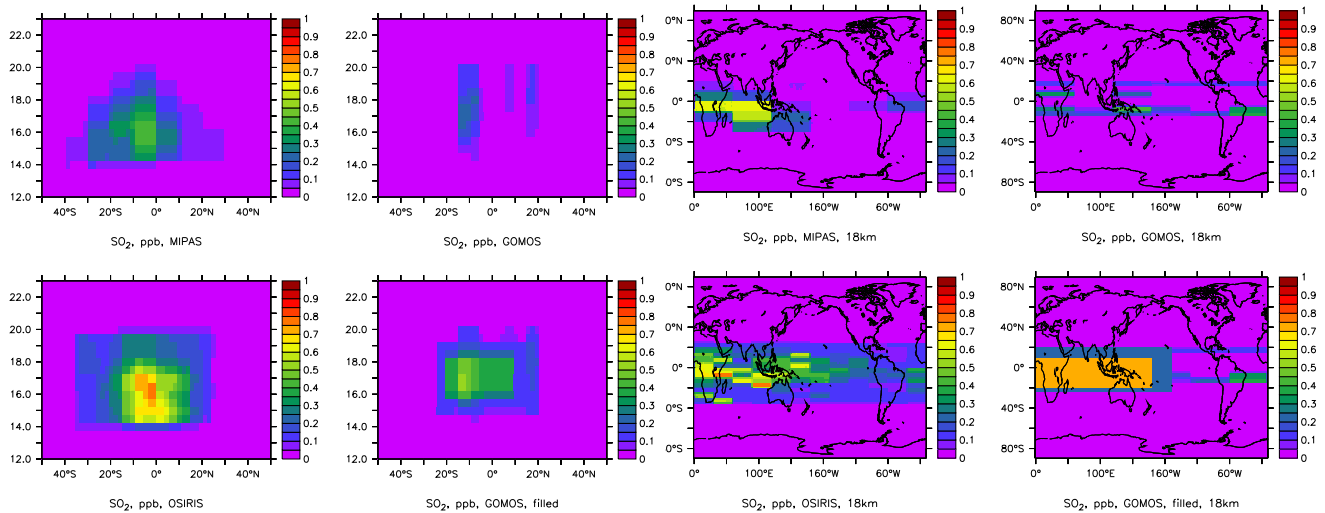
Figure B2. 2010 Merapi eruption: SO₂ derived from MIPAS, GOMOS OSIRIS and GOMOS with gap filling. Average vertical distribution (left) and plumes at 18 km (right).
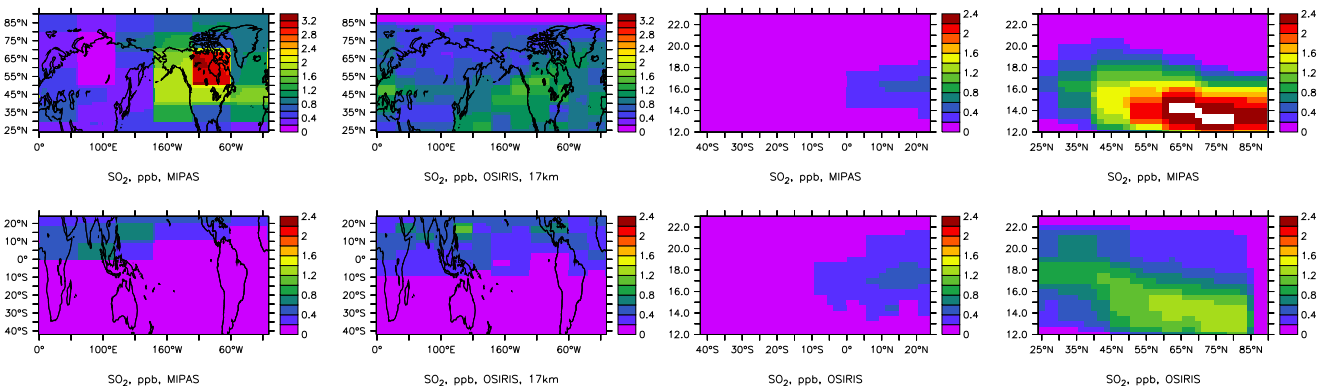
Figure B3. Sarychev and Mando Hararo from MIPAS and OSIRIS, arrangement of panels because event is split at 24° N

Appendix C: Comparison of different volcanic injection inventories

Year	Carn et al. (2016): total SO$_2$ (kt)	Carn et al. (2016): explosive SO$_2$ (kt)	in %	This study: stratospheric SO$_2$ (kt)
1990	186	186	100	744
1991	26082	24214	93	18424
1992	810	810	100	794
1993	450	450	100	426
1994	1874	360	19	337
1995	TOMS data gap	-	-	156
1996	987	100	10	436
1997	41	41	100	254
1998	3265	38	1.2	255
1999	130	85	65	398
2000	653	336	51	185
2001	1783	122	6.8	400
2002	2626	271	10	368
2003	679	679	100	207
2004	2997	410	14	256
2005	4634	2501	54	445
2006	1347	661	49	611
2007	712	122	17	450
2008	2625	2318	88	688
2009	1934	1379	71	839
2010	1470	867	59	424
2011	6030	4310	71	689
2012	763	563	74	355
2013	185	180	97	448
2014	5296	608	11	716
2015	-	-	-	993
2016	-	-	-	748
2017	-	-	-	600
2018	-	-	-	881
2019	-	-	-	1149

Table C1. Comparison of different volcanic emission inventories: Explosive and total annual amount of global volcanic SO$_2$ emissions and the percentage of explosive emissions, calculated from satellite observations in 1979 to 2014 (Carn et al., 2016), and the explosive volcanic SO$_2$ emissions reaching the stratosphere from the Volcanic Sulfur Emission Inventory (Table 2) in this study, ending in August 2019.

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