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## Assimilation of volcanic sulfur dioxide products from IASI and TROPOMI into the chemical transport model MOCAGE: case study of the 2021 La Soufrière Saint Vincent eruption with the March 2022 version of MOCAGE

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Abstract. Sulfur dioxide emitted during volcanic eruptions can be hazardous for aviation safety. As part of their activities, the Volcanic Ash Advisory Centres (VAACs) are therefore interested in the real-time atmospheric monitoring of this gas. A recent development aims at improving the forecasts of volcanic sulfur dioxide quantities made by the MOCAGE (Modèle de Chimie Atmosphérique à Grande Échelle) chemistry transport model. For this purpose, observations from both TROPOMI (Tropospheric Monitoring Instrument) and IASI (Infrared Atmospheric Sounding Interferometer; B and C) located on separate polar-orbiting satellites are assimilated into the model. These sulfur dioxide measurements are based on the eruption event of the La Soufrière Saint Vincent volcano in April 2021. Observations from OMI (Ozone Monitoring Instrument) are considered validation data. The resulting assimilation experiments show that the combined assimilation of IASI and TROPOMI observations always leads to a better forecast compared to the independent assimilation of data from each instrument. Sulfur dioxide atmospheric field forecasts are better when the available observations are numerous and cover a long time window.

## 1 Introduction

During volcanic eruptive events, large quantities of ash and sulfur dioxide  $(SO_2)$  are quickly released into the atmosphere. The emitted volcanic plumes can be transported far

from the emission sources, sometimes reaching the upper troposphere or even the stratosphere (Carn et al., 2009). At such altitudes, volcanic ash plumes become hazardous for aviation safety as they can irreversibly damage aircraft engines and significantly lower flight visibility (Prata, 2009). Aircraft passengers and crew are also directly threatened, especially because air quality inside and at the vicinity of volcanic plumes is strongly degraded, generating respiratory issues detrimental to human health (Schmidt et al., 2014).

Volcanoes can be monitored by in situ sensors except if they are difficult to reach and hazardous. Consequently, passive satellite remote sensing remains an efficient technique providing global data on gases and aerosols emitted during volcanic eruptions. Sulfur dioxide is one of the compounds measurable by remote sensing. The absorbing bands of this gas are in the ultraviolet (UV; ~ 310–340 nm) and thermal infrared (IR;  $-\nu_1 \sim 8.6 \mu m$ ,  $\nu_3 \sim 7.3 \mu m$  and  $\nu_1 + \nu_3 \sim 4 \mu m$ ) domains (Carn et al., 2016).

In many eruption cases like the one of the Icelandic volcano Eyjafjallajökull in 2010, volcanic sulfur dioxide can be considered a tracer to predict volcanic ash dispersion (Sears et al., 2013). However, both ash and sulfur dioxide plumes need to be monitored separately as their spatial distribution do not always coincide perfectly (Thomas and Prata, 2011). A striking example is the case of the eruption of the Icelandic volcano Grímsvötn in 2011, during which both sulfur dioxide and ash plumes were clearly separated (Prata et al., 2017).

Volcanic sulfur dioxide primary emissions can also be rapidly converted into secondary sulfate aerosols by reacting with water vapour and dioxygen. This conversion directly impacts the spatial distribution of volcanic sulfur dioxide plumes. The eruption of the Hunga Tonga volcano in January 2022 was exceptional as volcanic gases (sulfur dioxide and water vapour) and ash have been injected at an altitude of at least 30 km (Witze, 2022). As a result, stratospheric sulfur dioxide has been rapidly converted into sulfate aerosols because of water vapour propelled into the stratosphere during the submarine eruption (Sellitto et al., 2022).

To guarantee aviation safety, pilots need to dispose of accurate data on volcanic plume extent, movement and chemical composition. The International Civil Aviation Organization (ICAO) and the World Meteorological Organization (WMO) created the International Airways Volcano Watch (IAVW) in 1987 for this purpose (Lechner et al., 2018). Since 1990, the IAVW system has included nine worldwide responsibility areas, each represented by a Volcanic Ash Advisory Centre (VAAC). Europe, Africa and the Middle East are part of the Toulouse VAAC supervised by Météo-France (Gouhier et al., 2020).

Information on volcanic plumes provided by each VAAC to aviation authorities currently relies on specific services like the Support to Aviation Control Service (SACS) (Brenot et al., 2014) or the European Natural Airborne Disaster Information and Coordination System for Aviation (EUNADICS-AV) (Brenot et al., 2021). Based on in situ measurements, satellite data and modelling products, these systems provide VAACs information on volcanic ash and sulfur dioxide plumes. The Toulouse VAAC forecasts the dispersion of volcanic ash plumes by running MOCAGE-Accident (Gouhier et al., 2020), a specific version of the three-dimensional chemistry transport model (CTM) MOCAGE (Modèle de Chimie Atmosphérique à Grande Échelle) of Météo-France. To achieve this, an injection profile and a quantity of ash emitted by the volcano, previously chosen by the forecaster, are used to predict the dispersion of the volcanic plume.

Assimilation of volcanic sulfur dioxide observations into a model has already been performed in several situations like for the eruption events of Eyjafjallajökull in 2010 and Grímsvötn in 2011. Volcanic sulfur dioxide released by these volcanoes has been monitored by the GOME-2 (Global Ozone Monitoring Experiment-2) and OMI (Ozone Monitoring Instrument) UV sensors. The resulting retrieved observations have been assimilated in the Integrated Forecasting System for atmospheric composition (IFS-COMPO) of the European Centre for Medium-Range Weather Forecasts (ECMWF) and improved the sulfur dioxide plume forecasts (Flemming and Inness, 2013). Volcanic sulfur dioxide retrievals from the GOME-2 and TROPOMI (Tropospheric Monitoring Instrument) UV sensor observations have been operationally assimilated in the global IFS-COMPO assimilation system since October 2020. On top of that, a retrieved volcanic sulfur dioxide layer height from the TROPOMI Layer Height product has been assimilated. Including plume height information into the assimilation system enhanced the quality of the forecasts made from the analysed fields (Inness et al., 2022).

The eruption of the La Soufrière Saint Vincent volcano emitted a large amount of SO<sub>2</sub> into the atmosphere between 9 and 22 April 2021. According to the study of Esse et al. (2024), around 380 kt of SO2 was released into the atmosphere during the first 48 h of the eruption. This sulfur dioxide has been detected by several remote sensing instruments like the IR sensor IASI (Infrared Atmospheric Sounding Interferometer) and the UV sensors TROPOMI and OMI. The assimilation of TROPOMI, IASI and both instruments should improve MOCAGE. Indeed, without data assimilation, the model does not simulate the  $SO_2$  plume. In this study, we jointly assimilate a UV instrument, TROPOMI, and the IR instruments IASI B and IASI C, allowing for the correction of the model during both day and night. The use of instruments with different wavelengths to assimilate volcanic SO<sub>2</sub> data is also beginning to be developed in IFS. In MOCAGE, a part of the SO<sub>2</sub> can be converted into sulfate aerosols, particles which are causing problems in the aviation sector.

For the assimilation, the three-dimensional variational data assimilation system of the CTM MOCAGE (El Amraoui et al., 2022) is used. Forecasts of volcanic sulfur dioxide plumes are then initialised by the resulting analyses. Two preliminary experiments are conducted by independently assimilating total columns retrievals from the IASI and TROPOMI sensors. The resulting total columns of volcanic sulfur dioxide assimilated in MOCAGE are then compared to those measured by the OMI independent sensor, located on another satellite.

In this paper, the La Soufrière Saint Vincent eruption event of 2021 is introduced in the second part. Then, the assimilation data provided by the corresponding instruments are described in the third part, before the chemical transport model MOCAGE and its assimilation system in the fourth part. The fifth part addresses the results of this case study.

## 2 Description of the 2021 La Soufrière Saint Vincent eruption

La Soufrière Saint Vincent is a volcano located in the Grenadine Islands  $(13.33^{\circ} N, 61.18^{\circ} W)$ . The eruption in 2021 started on 9 April with a violent explosion around 12:40 UTC. This first explosion released a volcanic plume that reached an altitude of 8 km. As a result, thousands of people were forced to flee. A second and weaker explosion occurred at 18:45 UTC, generating a volcanic plume that reached an altitude of 4 km. At 22:35 UTC, a third explosion took place with a plume reaching 16 km. Between 10 and 11 April in the morning, the volcanic activity became periodic as explosions occurred every 1 to 3 h, during short time periods of 20 to 30 min each. Although the number of explosions decreased from 12 April, the volcanic plume re-

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mained at a high altitude, exceeding 12 km and sometimes even 16 km. Two last major explosions took place on 12 April at 08:15 UTC and on 13 April at 10:30 UTC with plumes reaching altitudes of 12.8 and 11 km respectively. The volcano continued to emit temporarily ash and volcanic gases in the atmosphere until 22 April, but the plume from these explosions no longer reached 8 km high. No fewer than 30 explosions were observed during this eruption event, most of them during 9 and 11 April. More information about the La Soufrière eruptions is available in the report of Bennis and Venzke (2021).

## **3** Description of the instruments

## 3.1 TROPOMI

TROPOMI is a hyperspectral radiometer with spectral bands extending from the UV to the shortwave infrared (SWIR) domains. This instrument is on board the polar-orbiting Sentinel 5 Precursor (S5p) satellite, whose goal is to provide information and services on air quality and climate (Reshi et al., 2024). Since August 2019, TROPOMI has benefited from a high spatial resolution of  $5.5 \text{ km} \times 3.5 \text{ km}$  at nadir. This instrument has a daily temporal resolution (no observation at night), and its local overpass time occurs at 13:35 UTC (Veefkind et al., 2012).

Volcanic sulfur dioxide plumes can be globally monitored with the high spatial resolution of TROPOMI. Unprocessed radiances measured by the instrument are often unpacked and formatted to become level 1 (L1) data. These data can then be converted into a proper retrieved environmental variable as sulfur dioxide, forming a level 2 (L2) product. This conversion requires the use of a retrieval algorithm having its own specificities and uncertainties. Historically, the first institutes studying sulfur dioxide L2 products with TROPOMI measurements were the Royal Belgian Institute for Space Aeronomy (BIRA) and the German Aerospace Center (Deutsches Zentrum für Luft- und Raumfahrt, DLR). For that, they use the differential optical absorption spectroscopy (DOAS) algorithm, particularly fitted for TROPOMI operational nearreal-time processing (Reshi et al., 2024).

Backscattered ultraviolet radiation is measured in order to construct absorption spectra. The DOAS algorithm is then applied to these spectra for different fitting windows between 310 and 390 nm. The DOAS algorithm operates over several steps. First, slant column densities (SCDs) are computed. They correspond to the integrated sulfur dioxide concentration along the mean atmospheric optical path. Then, conversion factors called air mass factors (AMFs) are obtained from suitable radiative transfer calculations to take measurement sensitivity changes into account. These changes depend on many factors like observation geometry, total ozone absorption, clouds and surface reflectivity. Moreover, the measurement sensitivity varies with the altitude of the emitted sulfur dioxide plume. As this altitude is unknown, the AMFs are computed for several hypothetical sulfur dioxide vertical profiles. One profile used for polluted scenarios comes from a forecast made by the TM5 chemical transport model (Huijnen et al., 2010). Three other profiles are available for 1 km thick boxes. The first box extends from the ground level to 1 km high. The two others are centred at 7 and 15 km above mean sea level. The first profile is located in the boundary layer and stands for well-mixed anthropogenic or volcanic sulfur dioxide conditions. The second profile aims at representing sulfur dioxide emitted by effusive volcanic eruptions in the upper troposphere. The third one is for sulfur dioxide released by explosive volcanic eruptions above the lower stratosphere (Theys et al., 2017). As four AMFs are available depending on different assumed SO<sub>2</sub> vertical profiles, the conversion of SCDs into vertical column densities (VCDs) generates four types of VCDs. These vertical columns correspond to the number of sulfur dioxide molecules in an atmospheric column per unit area, usually expressed in Dobson units  $(1 \text{ DU} = 2.69 \times 10^{16} \text{ molec. cm}^{-2})$ . Finally, averaging kernels are computed for the four vertical profiles (Theys et al., 2019).

For our study, we use sulfur dioxide total vertical columns computed from the hypothetical profile centred around 15 km. These columns are associated with their systematic errors in order to compute the observation error matrix of the MOCAGE assimilation system. Moreover, the averaging kernel matrix needs to be considered for comparing TROPOMI to other instrumental measurements or model calculations (Rodgers and Connor, 2003). Finally, data are selected according to the category of the TROPOMI detection. Many cases are taken into consideration in our study: flag 1 for sulfur dioxide detection, flag 2 for clear volcanic detection and flag 3 for detection close to a known anthropogenic source. The quality of the SO<sub>2</sub> retrieval is given by a quality flag with values ranging from 0 for uncertain retrieval to 1 for the best retrieval. TROPOMI data are available on the NASA website (https://disc.gsfc.nasa.gov/, last access: 19 September 2024).

## 3.2 TROPOMI Layer Height product

The TROPOMI Layer Height product (Hedelt et al., 2019) allows for determining the altitude of an SO<sub>2</sub> plume when TROPOMI SO<sub>2</sub> total columns are higher than 20 DU thanks to a machine learning algorithm called the Full-Physics Inverse Learning Machine (FP\_ILM). Hedelt et al. (2019) used the LInearized Discrete Ordinate Radiative Transfer (LIDORT) model (Spurr et al., 2008) to simulate many reflectance spectra for different values of the solar zenith angle (SZA), viewing zenith angle (VZA), relative azimuth angle (RAA), O<sub>3</sub> and SO<sub>2</sub> vertical column density, layer height, surface albedo, and surface pressure. Before separating these reflectance spectra into 10 principal components (PCs), the TROPOMI spectral response function, character-

ising the sensitivity of the instrument across its measurement spectrum, is applied. These PCs and information about the surface,  $O_3$  total columns, SZA, VZA and RAA are used as input for the neural network. SO<sub>2</sub> total columns and SO<sub>2</sub> height are diagnosed thanks to the neural network. In this study, SO<sub>2</sub> total columns diagnosed by the neural network are not assimilated. Nevertheless, the SO<sub>2</sub> height product is used to validate the altitude of the modelled plume.

TROPOMI Layer Height data are available on the NASA website (https://disc.gsfc.nasa.gov/, last access: 12 April 2024). Older data were provided by the German Aerospace Center (DLR).

## 3.3 IASI

IASI is a Fourier transform spectrometer operating in the IR spectral domain. This instrument is located on both polarorbiting MetOp-B (IASI B) and MetOp-C (IASI C) satellites. The best spatial resolution at nadir is a circle with a diameter of 12 km. Twice a day (measurements are possible both during daytime and nighttime), the IASI instruments observe at around 09:30 and around 21:30 local time (LT) (Clerbaux et al., 2009).

Sulfur dioxide observation data provided by IASI measurements are converted into a level 2 product using the ULB-LATMOS retrieval algorithm (Clarisse et al., 2012). In our study, we use an optimal sulfur dioxide total vertical column computed from an estimated altitude of the volcanic plume. This estimation is based on another algorithm created for IASI sulfur dioxide plume altitude retrievals (Clarisse et al., 2014). IASI data are available at the AERIS data centre (https://iasi.aeris-data.fr/, last access: 19 September 2024).

## 3.4 OMI

OMI is a multispectral radiometer with spectral bands extending from the UV to the visible (VIS) domains. This sensor is carried by the polar-orbiting Earth Observing System (EOS) Aura satellite. The best nadir spatial resolution is about  $24 \text{ km} \times 13 \text{ km}$ . Since 2011, this instrument has had 2 d daily coverage with an overpassing at 13:45 LT (Qu et al., 2019).

OMI sulfur dioxide total vertical columns are used as independent observations to check the results of our assimilation experiments. These total vertical columns are retrieved thanks to an algorithm based on a principal component analysis (PCA) technique (Li et al., 2017). These vertical columns are computed by considering a hypothetical sulfur dioxide plume altitude located around 18 km. OMI data are available on the NASA Earthdata website (https://omisips1.omisips.eosdis.nasa.gov/outgoing/ OMSO2NRTb/, last access: 19 September 2024).

#### 4 Description of the SO<sub>2</sub> assimilation experiments

MOCAGE (Modèle de Chimie Atmosphérique à Grande Échelle) is the CTM developed by the Centre National de Recherches Météorologiques (CNRM) at Météo-France (Josse et al., 2004). It has many operational uses such as air quality forecasting over France (Rouil et al., 2009) and over Europe, contributing data to the CAMS ensemble forecasting system (Marécal et al., 2015). MOCAGE is also used in a configuration without chemical reactions by the Volcanic Ash Advisory Centre of Toulouse (VAAC) when a volcanic eruption or an industrial accident occurs.

## 4.1 The model and its assimilation system

The CTM MOCAGE is a model using a semi-Lagrangian scheme for the transport of chemical species which can be global or nested. It enables predicting chemical evolution of the atmosphere up to 4 d. In this study, we use MOCAGE on a 1° global domain with 47 hybrid  $\sigma$  pressure levels distributed between the surface and 5 hPa (7 in the planetary boundary layer, 20 in the free troposphere and 20 in the stratosphere).

MOCAGE is an offline model and needs meteorological fields like wind speed and direction, temperature, humidity, pressure, rain, and clouds from a numerical weather prediction (NWP) model or from a climate model. In this study, meteorological forcings are provided by the French NWP model Action de Recherche Petite Echelle Grande Echelle (ARPEGE) (Courtier et al., 1991; Bouyssel et al., 2022).

The model enables transforming species according to the chemical scheme RACMOBUS, which is a combination of two chemical schemes. The first one, RACM, is computed for tropospheric chemical reactions (Stockwell et al., 1997). It is completed with the sulfur cycle (Feinberg et al., 2019). The second chemical scheme, REPROBUS, is used for stratospheric chemical reactions (Lefevre et al., 1994). Every 15 min, the MOCAGE model provides the atmospheric composition of 112 gaseous species thanks to 379 chemical reactions and 57 photolysis reactions.

Both primary and secondary aerosols are modelled in MOCAGE (Guth et al., 2016; Sič et al., 2015). Primary aerosols include desert dust, sea salt, black carbon, organic carbon and volcanic ash. In this study, volcanic ash modelling is turned off. Secondary inorganic aerosols are represented by sulfate, nitrate and ammonium aerosols. The aerosol size distribution is described by a sectional approach, with six size sections delimited by the following diameters: 0.002-0.01, 0.01-0.1, 0.1-1.1, 1.1-2.5, 2.5-10 and  $10-50 \,\mu$ m. Desert dust and sea salt emissions depend on the wind strength and the type of the ground.

To forecast air quality, emissions of gaseous and aerosol species need to be taken into account by the model. Emission inventories are therefore used, such as the MACCity inventory for anthropogenic emissions (Lamarque et al., 2010) and the MEGAN inventory for biogenic emissions (Sinde-

larova et al., 2014). Sulfur dioxide released into the atmosphere by passive degassing can also be, as in our study, part of the emissions included in the model (Lamotte et al., 2021). Daily emissions of biomass burning provided by the Global Fire Assimilation System (GFAS) (Kaiser et al., 2012) are injected into the model at different vertical levels, depending on the latitude of fires (Cussac et al., 2020). Biomass burning emissions are injected at an altitude of 1 km in the tropics, 2 km at middle latitudes and 6 km at high latitudes. Other species except lightning nitrogen oxides (NO<sub>x</sub>) (Price et al., 1997) and aircraft emissions (Lamarque et al., 2010) are emitted in the first five levels of the model (approximately 500 m altitude).

Many products can be assimilated in MOCAGE. For example, to improve  $O_3$  in MOCAGE, total columns (Emili et al., 2014) or radiances (El Aabaribaoune et al., 2021; Vittorioso et al., 2024) can be assimilated. For the aerosols, aerosol optical depth (AOD) (Sič et al., 2016; El Amraoui et al., 2022) or lidar observations (El Amraoui et al., 2020; Cornut et al., 2023) can be assimilated into MOCAGE.

The assimilation system used in this study is 3D-VAR (three-dimensional variational assimilation), described hereafter. A short-range forecast from MOCAGE  $x^b$  and observations y are combined to find the optimal state  $x^a$ , taking into account their respective error covariance matrices **B** and **R**.  $x^a$  is the sum of  $x^b + \delta x^a$ , where  $\delta x^a$  is the increment minimising the cost function J:

$$J(\delta x) = \frac{1}{2} (\delta x)^T \mathbf{B}^{-1} (\delta x) + \frac{1}{2} \left( y - \mathcal{H} \left[ x^b \right] - \mathcal{H} \left[ \delta x \right] \right)^T$$
$$\mathbf{R}^{-1} \left( y - \mathcal{H} \left[ x^b \right] - \mathcal{H} \left[ \delta x \right] \right), \tag{1}$$

where  $\mathcal{H}$  is the observation operator used to obtain the model data in the observation space. Before running an assimilation experiment, a full description of the **R** and **B** matrices is required. The background error covariance is spread in space thanks to the correlation matrix described in El Amraoui et al. (2020). This matrix contains both horizontal and vertical components.

The horizontal correlation  $C_{m,n}^{h}$  between two points *m* and *n* is defined as follows:

$$C_{m,n}^{\rm h} = \exp\left[\frac{-d^2}{2\left(L_{x^2} + L_{y^2}\right)}\right],$$
 (2)

where *d* is the distance between the points *m* and *n* and  $L_x$  and  $L_y$  are the longitude and latitude length scales in kilometres. In our study, the longitude and latitude length scales are equal to one mesh grid (1°).

In kilometres, the length scales become

$$L_x = L_y = 2R_e \cdot \sin\left(\frac{\pi}{360}\right),\tag{3}$$

where  $R_e$  is Earth's radius (6371.22 km).

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**Table 1.** Description of the experiments performed during this study with the different assimilated instruments.

Experiment	Assimilation of TROPOMI	Assimilation of IASI B and C	Assimilation of OMI
iasi_assim	No	Yes	No
tropomi_assim	Yes	No	No
joint_assim	Yes	Yes	No
dry	No	No	No

The vertical correlation  $C_{i,j}^{v}$  between two pressure levels  $(p_i \text{ and } p_j)$  is defined as follows:

$$C_{i,j}^{\mathsf{v}} = \exp\left[-100 \cdot \left[\log\left(\frac{p_i}{p_j}\right)\right]^2\right].$$
(4)

In our study, the values of the vertical correlation between two consecutive levels are set to 1.

## 4.2 TROPOMI and IASI data assimilation setup

Several hourly 3D-VAR assimilations of volcanic SO<sub>2</sub> data into MOCAGE have been conducted over the specific eruptive period from 9 to 15 April 2021 with a 1° horizontal resolution. Different simulations have been carried out: one with the assimilation of IASI B and C (iasi\_assim), one with the assimilation of TROPOMI (tropomi\_assim), another with the assimilation of IASI and TROPOMI, and the last one without assimilation (Table 1). The results of these experiments are compared to OMI observations.

In these experiments, IASI and OMI observations above 0.5 DU are used. This value corresponds to the lowest total columns measurable by these instruments (Koukouli et al., 2022; Qu et al., 2019). For TROPOMI, observation retrievals with an SO<sub>2</sub> peak concentration at 15 km altitude are used. Moreover, an observation is used when the quality flag is above 0.5 and if the slant column is above 1 DU, matching the noise of the instrument.

Averaging kernels are taken into account in this study. For TROPOMI, averaging kernels are only given for the a priori profiles from the TM5 CTM. Nevertheless, averaging kernels for the other a priori profiles can be estimated by multiplying them by a scaling factor (Theys, 2018) as described by the following equation:

$$AVK(z) = \frac{AVK_{TM5}(z) \times AMF_{TM5}(z)}{AMF_{15\,km}},$$
(5)

where AVK(z) is the averaging kernels at a given altitude, AVK<sub>TM5</sub>(z) and AMF<sub>TM5</sub>(z) are the averaging kernels and the air mass factor at altitude z from the TM5 CTM, and AMF<sub>15 km</sub> is the air mass factor for the a priori profile containing a peak at 15 km altitude.

For IASI, averaging kernels are not given in the observation files. However, they can be computed at many heights



**Figure 1.** Values of the observation error standard deviation (sigR) according to TROPOMI (**a**) and IASI (**b**) observations.

thanks to the  $SO_2$  total columns, according to the following equation:

$$AVK(z) = \frac{Y(z)}{Y(z_{ref})},$$
(6)

where z is the hypothetical SO<sub>2</sub> injection altitude, AVK(z) is the averaging kernels at a given altitude z, Y(z) is the total columns computed for an SO<sub>2</sub> injection at altitude z and  $Y(z_{ref})$  is the total column computed for a reference altitude injection. In our case, this altitude is provided by the observation files.

In this study, we assume there is no spatial correlation in the observation error. For TROPOMI, the observation error standard deviation (sigR) is set according to the uncertainties provided by the instrument for each observation (Fig. 1a). The observation error standard deviation is set to around 25 % of the observations for TROPOMI. The uncertainties in IASI measurements vary according to the value of the total column measured. In the case of this eruption, IASI measured total SO<sub>2</sub> columns ranging from 0.5 to 20 DU. The uncertainties in this range of observations vary from around 25 % to 5 % of the observation (Clarisse et al., 2012). For IASI, we set the observation error standard deviation to 15 % of the observation values (Fig. 1b).

For observations greater than 20 DU, the TROPOMI Layer Height product is able to diagnosed the altitude of the plume. During this volcanic eruption, 90 % of the diagnosed heights are between 9 and 21 km. Consequently, to force SO<sub>2</sub> injection between these two altitudes, we chose a profile containing strong values  $(1 \times 10^{-8} \text{ ppv})$  between altitudes of 9 and 21 km as the background error standard deviation. It is important to note that this setting of the background error standard deviation is only valid for this volcanic eruption and that, for another eruption, the user must choose a different profile of the background error standard deviation to inject  $SO_2$  at the correct altitude into the model. The correlation matrix is the same in all experiments with data assimilation.

As in the operational mode of MOCAGE, a forecast initialised by the assimilation outputs is launched at 00:00 UTC each day. In our study, this forecast is performed for up to a 72 h term range.

### 5 Impact on analyses

#### 5.1 Impact on SO<sub>2</sub> and sulfate

Figure 2 shows the SO2 total columns observed and analysed on 10 April 2021 at various times (02:00, 14:00, 16:00 and 17:00 UTC). The observations are depicted on the first line for TROPOMI and the second and the third lines for IASI B and C respectively. The model's outputs are on the fourth line for the iasi assim experiment, on the fifth line for the tropomi assim experiment and on the sixth line for the joint\_assim experiment. At 02:00 UTC, the tropomi\_assim simulation does not present an SO<sub>2</sub> plume, in contrast to the plume modelled with IASI assimilation. Nevertheless, the model underestimates the total column values as compared to the IASI observations. Due to the use of UV wavelengths, TROPOMI is unable to measure SO<sub>2</sub> total columns at night. Consequently, no SO<sub>2</sub> plume is modelled in MOCAGE until 17:00 UTC. Before the overpass of TROPOMI, IASI instruments measure SO<sub>2</sub> total columns once again. It allows for increasing both the intensity and size of the plume in MOCAGE at 14:00 UTC. At 17:00 UTC, a plume appears in the tropomi assim experiment thanks to the TROPOMI overpass. Compared to the TROPOMI observations, values of high total columns are underestimated by MOCAGE. At this time, the simulated volcanic plume in the iasi\_assim experiment is slightly smaller and weaker than the plume in the tropomi assim experiment and the TROPOMI observations. Assimilation of TROPOMI data allows for simulating a strong area value in the vicinity of the volcano in both the tropomi\_assim and joint\_assim experiments. This structure of strong values is not modelled with the IASI data assimilation because it corresponds to the latest volcanic SO<sub>2</sub> emission. This new release, due to a new eruption event, took place between the IASI and the TROPOMI overpasses. Simulated SO<sub>2</sub> total columns in the joint\_assim experiment are stronger, in particular around 55° W, where observations are strong. For this part of the plume, the difference between the model and the observations is weaker because it is analysed five times during this day, whereas the model is corrected four times when IASI is assimilated and only once when TROPOMI is assimilated. The greater the number of model corrections, the smaller the differences between observations and the model. In this experiment, the shape of the plume is slightly different with a pattern around 39° W, which is simulated with the IASI data assimilation and not with the TROPOMI one.



**Figure 2.** Observations assimilated and analyses of  $SO_2$  total columns on 10 April 2021 at 02:00, 14:00, 16:00 and 17:00 UTC. The first three rows correspond to TROPOMI, IASI B and IASI C observations respectively. Analysis outputs are plotted on the fourth line for the iasi\_assim experiment, on the fifth line for the tropomi\_assim experiment and on the sixth line for the joint\_assim experiment. The shaded areas on the first three lines correspond to areas where there are no observations or where observations have not been assimilated. Observations are not assimilated when they are less than 0.5 DU for IASI and TROPOMI's slant columns are less than 1 DU.

Figure 3 shows the  $SO_2$  total columns observed and analysed on 11 April 2021 at 11:00, 13:00, 17:00 and 18:00 UTC. Until but not including 17:00 UTC, the  $SO_2$  plumes simulated by the iasi\_assim and the joint\_assim experiments are similar. At 11:00 UTC, the eastern parts of the plume are con-

sistent between experiments. Thanks to the IASI overpass at the end of the previous day, the plume is closer to the volcano and  $SO_2$  total columns of the western part of the plume are stronger compared to the tropomi\_assim experiment. At 13:00 UTC, IASI SO<sub>2</sub> total columns are assimilated, reducing the SO<sub>2</sub> plume over the ocean and increasing the values in the vicinity of the volcano in the model. At 17:00 UTC, TROPOMI observations are assimilated and the shape of the SO<sub>2</sub> plume is consistent between the experiments. The SO<sub>2</sub> plume is more extended with the assimilation of TROPOMI. Concerning the values of the total columns, areas with high values are more intense regarding the assimilation of both instruments, particularly near the volcano and in the middle of the Atlantic Ocean. At 18:00 UTC, the OMI overpass enables validating these experiments with independent observations. The SO<sub>2</sub> plume simulated by the joint\_assim experiments seems to be the closest to OMI observations. Indeed, in this experiment, the strong SO<sub>2</sub> total column patterns match better to the observations, in particular around 39 and around  $55^{\circ}$  W.

The vertical cross-section at 13.5° N, illustrated in Fig. 4, presents the vertical SO<sub>2</sub> concentrations at various times on 11 April 2021 (11:00, 13:00, 17:00 and 18:00 UTC). The chosen assimilation settings result in a plume that extends from 9 to 21 km in altitude. Until but not including 17:00 UTC, the altitude of the maximum SO<sub>2</sub> concentration in the different experiments is consistent with an altitude around 13 to 17 km. The lowest concentrations are simulated in the tropomi\_assim experiment. At 11:00 UTC, the IASI assimilation of the end of the previous day enables simulating a plume located closer to the volcano compared to the tropomi\_assim experiment. At 13:00 UTC, with the assimilation of IASI, a new strong concentration pattern appears near the volcano around 13 km altitude. At 17:00 UTC, in experiments where TROPOMI observations are assimilated, SO2 concentration increases around 9 km altitude in the vicinity of the volcano. The altitude of the plume is closer to the TROPOMI Layer Height product in the joint\_assim experiment. However, around 50° W, the TROPOMI Layer Height product shows a plume between 10 and 16 km altitude. In the experiments, the height of the plume varies between 15 and 20 km, which is too high compared to the TROPOMI Layer Height product. Nevertheless, around 50° W, few observations meet the criteria for the TROPOMI Layer Height product to be able to diagnose the height of the plume. It is difficult to conclude whether the plume altitude is correctly represented in the model when there are few or no observation above 20 DU.

 $SO_2$  can be converted into sulfate aerosol in the presence of water vapour. This process is modelled in MOCAGE. Indeed, sulfate total columns show structures in the assimilation experiment which are not shown without assimilation.

Figure 5 shows the SO<sub>2</sub> total columns in the top panel, the sulfate total columns in the middle panel and the aerosol optical depth (AOD) in the bottom panel on 14 April 2021 at 07:00 UTC for different experiments with assimilation (iasi\_assim, tropomi\_assim and joint\_assim) and without assimilation (dry). This figure shows strong differences between each experiment. Indeed, on this date, the analysed SO<sub>2</sub> total columns are stronger by assimilating

TROPOMI than in the iasi\_assim and the joint\_assim experiments. Nevertheless, SO<sub>2</sub> total column values never reach 3 DU. Without assimilation, no SO<sub>2</sub> plume is modelled by MOCAGE.

The assimilation of volcanic SO<sub>2</sub> total columns allows the model to simulate sulfate aerosols (middle panel of Fig. 5). Indeed, a sulfate plume is simulated from the volcano to Guinea. This sulfate plume is not modelled in the experiment without assimilation. In the tropomi\_assim experiment, strong sulfate total columns are simulated in French Guiana and in Venezuela. In this area, values exceed 25 mg m<sup>-2</sup>, whereas values do not reach 20 mg m<sup>-2</sup> in the iasi\_assim and joint\_assim experiments. Elsewhere the modelled sulfate total columns are consistent with from one assimilation experiment to another.

Total AOD values slightly increased in Central America with the SO<sub>2</sub> assimilation (fifth line in Fig. 5). This rise is more important regarding assimilating TROPOMI in French Guiana and in Venezuela, where AOD values reach 0.5 compared to 0.4 in the iasi\_assim and in the joint\_assim experiments and 0.3 without assimilation (fourth line in Fig. 5). However, few AOD observations from the MODIS (Moderate-Resolution Imaging Spectroradiometer) and VI-IRS (Visible Infrared Imaging Radiometer Suite) instruments are available in this area. This makes it impossible to validate AOD assimilation.

The assimilation of SO<sub>2</sub> total column measurements significantly enhances MOCAGE's ability to describe SO<sub>2</sub> plumes. When no observations are assimilated, no SO<sub>2</sub> plume is represented by the model. However, when satellite observations are assimilated, an SO<sub>2</sub> plume is simulated and corrected more or less frequently depending on the instrument or combination of instruments and thus on the overpass time of the corresponding satellites.

# 5.2 Impact of the assimilation on the detection of SO<sub>2</sub> threshold exceedances

To assess the impact of assimilation on volcanic SO<sub>2</sub>, the number of grid cells reaching the 1 and 5 DU thresholds, over a sub-domain extending from 90° W to 40° E and from 20° S to 30° N, has been calculated and plotted in Fig. 6 for the iasi\_assim (in red), tropomi\_assim (in blue) and joint\_assim (in green) simulations and for the TROPOMI (in orange), IASI B (in purple), IASI C (in magenta) and OMI (in grey) observations. Since there are often several observations per grid cell, we looked at the number of grid cells where the minimum and maximum of the total columns exceed these thresholds. These values are represented by horizontal lines. The number of grid cells where the median of the observations is higher than these thresholds is shown by a dot. Finally, the limits of the bars represent the number of grid cells where the 25th and 75th quantiles exceed the thresholds.



**Figure 3.** Observations and analyses of  $SO_2$  total columns on 11 April 2021 at 11:00, 13:00, 17:00 and 18:00 UTC. The first three rows correspond to TROPOMI, IASI B and IASI C observations respectively. The last row corresponds to OMI observations. Analysis outputs are plotted on the fourth line for the iasi\_assim experiment, on the fifth line for the tropomi\_assim experiment and on the sixth line for the joint\_assim experiment.



**Figure 4.** Vertical sections of the analysed SO<sub>2</sub> concentration at  $13.5^{\circ}$  N on 11 April 2021 at 11:00, 13:00, 17:00 and 18:00 UTC. Rows correspond to the TROPOMI data assimilation, IASI assimilation, joint assimilation and height of the SO<sub>2</sub> plume provided by the TROPOMI Layer Height product respectively.

Between 9 and 11 April 2021, the number of meshes where the median of the observations exceeded 1 DU is consistent between instruments. After this date, there were more grid cells where the median number of observations exceeded 1 DU with TROPOMI. Moreover, with this instrument, the dispersion of observations is high. There are few grid cells where all the observations exceed 1 DU, whereas on 14 April, there were more than 1200 grid cells where the maximum number of observations reaches 1 DU. This dispersion is lower for instruments with lower resolutions. For IASI, the number of grid cells where the median of observations is greater than or equal to 1 DU varies less than with UV instruments. This can be explained by IASI's high sensitivity to water vapour, which masks part of the SO<sub>2</sub> column. In the assimilation experiments, the number of points where the total column reaches 1 DU is identical between iasi\_assim and joint\_assim until the end of the day on 10 April 2021. Until this date, the number of grid cells where the total column reaches 1 DU is zero with the TROPOMI assimilation. After this date, the number of grid cells where the total column exceeds 1 DU is always bigger in the joint\_assim experiment. The number of grid cells above 1 DU is lower when only TROPOMI is assimilated in the morning until 11 April. This number is smaller when only IASI is assimilated. The differences in the number of grid cells exceeding 1 DU are greater at the end of the study period. Until 11 April 2021 in the experiments where TROPOMI was assimilated and until 12 April 2021 with the assimilation of IASI alone, the number of grid cells where

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**Figure 5.** SO<sub>2</sub> total column, sulfate total column, AOD, and the difference between AOD and the AOD of the dry experiment on 14 April 2021 at 7:00 UTC for the dry, iasi\_assim, tropomi\_assim and joint\_assim experiments.

the model exceeded 1 DU was consistent with the number of grid cells where the median of the observations exceeded this threshold. After this date, the number of grid cells exceeding 1 DU exceeds the number of meshes where the 75th quantile of TROPOMI observations exceeds 1 DU. Whatever the instruments used, the number of points above 1 DU is too high compared to IASI and OMI observations. This shows that the extension of the SO<sub>2</sub> plume is too large in the model.

The number of grid cells with observations exceeding 5 DU is lower and similar for each instrument. On 9, 14 and 15 April 2021, none of the observations exceeded this threshold. In the model, no column exceeds 5 DU for these dates. Generally, the number of  $SO_2$  columns exceeding 5 DU in the model is similar to the number of grid cells where the median of observations exceeds 5 DU. Between the end of the day on 10 and 12 April, the number of meshes in MOCAGE is slightly greater by assimilating the two instruments. Generally, this number is closer to the number of meshes where the median of the OMI observations reaches 5 DU except at the end of the day on 10 April 2021, when the number of meshes above 5 DU is slightly underestimated. On this day, this number is underestimated in the model because a new eruption took place between the last assimilation of TROPOMI and

the overpass of OMI. On 11 April, the TROPOMI assimilation added a significant number of points above 5 DU in the model because of a large number of TROPOMI observations above 5 DU. On 12 April, the extension of the plume reaching 5 DU is greater when assimilated in the tropomi\_assim experiment. In fact, the value of the total columns falls in the model thanks to the assimilation of IASI. The TROPOMI overpass at the end of the afternoon also reduces the modelled total columns. The number of points above 5 DU in the model becomes similar.

To assess the accuracy of the model in simulating  $SO_2$  total columns, a threshold-based analysis was implemented. The goal was to determine the number of instances where both the observations and the model successfully identified  $SO_2$  total columns above certain thresholds (labelled as hits), instances where the observations exceeded these thresholds but the model failed to detect them (labelled as misses), instances where the model exceeds these thresholds but the observations do not reach these thresholds (labelled false alarms), and instances where both the observations and the model successfully identified  $SO_2$  total columns under certain thresholds (labelled as correct rejections). Using these numbers, we defined three metrics.



Figure 6. Number of grid cells where analyses exceed 1 and 5 DU. The blue, red and green lines show the number of points at which the total columns reach these thresholds in the tropomi\_assim, iasi\_assim and joint\_assim experiments. Orange, purple, magenta and grey boxplots represent the number of grid cells where TROPOMI, IASI B, IASI C and OMI observations exceed the threshold.

The first one is the probability of detection (POD), a ratio that ranges from 0 to 1. The POD is calculated by dividing the number of hits by the sum of hits and misses for a given threshold. A POD score of 1 indicates a perfect detection by the model, meaning that all observed instances above the threshold were correctly simulated. On the other hand, a POD of 0 signifies that none of the observed SO<sub>2</sub> total columns above the threshold were detected by the model. The POD is computed with the following equation:

$$POD = \frac{hits}{hits + misses}.$$
 (7)

The second one is the critical success index (CSI), a ratio that ranges from 0 to 1. The CSI is calculated by dividing the number of hits by the sum of hits, misses and false alarms for a given threshold. A CSI score of 1 indicates perfect detection by the model, meaning that all observed instances above the threshold were correctly simulated. On the other hand, a POD of 0 signifies that none of the observed  $SO_2$  total columns above the threshold were detected by the model. The CSI is computed with the following equation:

$$CSI = \frac{hits}{hits + misses + false alarms}.$$
 (8)

The last one is the false alarm rate (FAR), a ratio that ranges from 0 to 1. The FAR is calculated by dividing the number of false alarms by the sum of false alarms and correct rejections for a given threshold. A FAR score of 0 indicates that there are only correct rejection instances. On the contrary, a FAR score of 1 indicates that there are only false alarm instances. The FAR is calculated with the following equation:

$$FAR = \frac{\text{correct rejections}}{\text{correct rejections} + \text{false alarms}}.$$
(9)

To study these metrics, the notations in Table 2 are adopted. For POD metrics, times when there are neither hits nor misses are shown by a dot and times when there are no hits but there are misses are represented by a cross. For CSI metrics, times when there are no hits, no misses and no false alarm are shown by a dot and times when there are no hits and no false alarms but misses occur are represented by a cross. If there are no hits but if there are misses and false

cross. If there are no hits but if there are misses and false alarms, a star is plotted. For FAR metrics, a dot is plotted when there are no false alarms and no correct rejections. A cross is plotted when there are no false alarms but there are correct rejections.

Figure 7 shows the probability of detection computed for the 1 and 5 DU thresholds against TROPOMI and IASI observations. Dots represent times when there is no observation. Crosses represent the moments when simulated  $SO_2$ total columns are under a threshold, whereas some observations exceeds this threshold. Compared to TROPOMI, POD values are generally better in the experiments in which TROPOMI observations have been assimilated. In these experiments, POD values exceed 0.75. The POD values are over 0.75 until 12 April in the iasi\_assim experiment except on 9 April. POD values decrease at the end of the study period. For the 5 DU threshold, POD values are slightly higher in the joint\_assim experiment, especially on 10 and 11 April, when around 100 TROPOMI observations exceed 5 DU. On 12 April, POD values are around 0.4 in the experiments in which TROPOMI is assimilated. No SO<sub>2</sub> total column higher than 5 DU is simulated for this date in the iasi\_assim experiment. Between 13 and 15 April and on 9 April, no SO<sub>2</sub> total column above 5 DU is simulated in MOCAGE. For these days, between one and nine observations above 5 DU are measured by TROPOMI.

POD values, computed for a 1 DU threshold and with IASI observations, exceed 0.75 in experiments in which IASI instruments are assimilated. No SO<sub>2</sub> total columns are simulated with the TROPOMI assimilation until 10 April because TROPOMI overpasses the plume after IASI. On the morning of 9 April, no observation above 1 DU is detected by IASI. From 11 April, POD values vary between 0.3 and 0.8 in the tropomi\_assim experiment. In this experiment, POD values are often higher in the morning. In the afternoon of 9 April, only one observation above 5 DU is measured by IASI. At this location, the total column is under 5 DU. For this threshold and compared to the tropomi\_assim experiment, the probability of simulating high increases in SO<sub>2</sub> total columns thanks to the IASI assimilation. Despite numerous observations above 5 DU, many events are missed on 10 April with a POD reaching nearly 0.4 in the morning and 0.5 in the afternoon. Most of the simulated SO<sub>2</sub> total columns are between 1 and 5 DU. The maximum POD is obtained on 11 April after the assimilation of many observations measured by IASI exceeding 5 DU on 10 and 11 April.

The first line in Fig. 8 shows the POD computed against OMI observations for the 1 and 5 DU thresholds. On this

line, dots represent times when there are no hits and no misses. Crosses represent the moments when simulated SO<sub>2</sub> total columns are under a threshold, whereas some observations exceed this threshold. The POD values computed with a 1DU threshold are often consistent between the tropomi\_assim and the joint\_assim experiments. POD values are slightly better in the joint\_assim experiment between 9 and 13 April. For the 5 DU threshold, POD value is greater with the joint\_assim experiment on 10 and on 11 April. On 12 and 13 April, no SO<sub>2</sub> total column above 5 DU is modelled by MOCAGE, whereas OMI measured observations above this threshold. Elsewhere in the study period, no observation greater than 5 DU is measured by OMI and modelled by MOCAGE.

The second line in Fig. 8 shows the CSI computed against OMI observations for the 1 and 5 DU thresholds. As for the POD, the CSI values computed with a 1 DU threshold are often consistent between the tropomi assim and the joint assim experiments. CSI values are slightly better in the joint\_assim experiment on 9, 11 and 12 April. On 10 and 11 April, CSI values are around 0.75, whereas POD values are around 0.9. It means that there are few false alarms during this both days. On the contrary, from 13 April, CSI values are much lower than POD values. It means that the plume in MOCAGE becomes too large, leading to a lot of false alarms. For the 5 DU threshold, CSI values are better in the joint\_assim experiment on 10 April and to a lesser extent on 11 April. From 14 April, no observations higher than 5 DU are measured by OMI and modelled by MOCAGE. On 9 and 12 April with the tropomi\_assim experiment and on 13 April, there are no OMI observations greater than 5 DU but some values above 5 DU are simulated by MOCAGE. On 12 April, there are misses and false alarms in the iasi\_assim and joint assim experiments.

The third line in Fig. 8 shows the FAR computed against OMI observations for the 1 and 5 DU thresholds. The FAR values computed with a 1 DU threshold are similar between experiments. Up to 11 April, FAR values are approximatively equal to 0. This shows that the number of correct rejections is larger than the number of false alarms. The FAR values increases from 12 April, meaning that the plume is too large in the model. The FAR values computed with the 5 DU threshold are always close to 0. On 9 and 12 April in the tropomi\_assim experiment and from 13 April, there are only correct rejections.

In the various modelling experiments, assimilating  $SO_2$  total column data enhances the performances of the CTM MOCAGE by enabling the simulation of an  $SO_2$  plume. However, for the lowest concentration threshold, the simulated  $SO_2$  plume tends to be overly extensive and the corresponding  $SO_2$  burden is too high when compared to observational data. Nevertheless, for higher thresholds, the  $SO_2$  plume area and  $SO_2$  burden are consistent with the observations in the experiments where TROPOMI is assimilated. Especially for the strong thresholds, POD values show an im-



Table 2. Symbols used in the plots according to the studied metric and the number of hits, misses, false alarms and correct rejections.







**Figure 7.** Probability of detection for the 1 and 5 DU thresholds for the three experiments: tropomi\_assim in blue, iasi\_assim in red and joint\_assim in green. Dots represent times when there are no observations. Crosses represent the moments when there are only misses. AM: morning, PM: afternoon.

provement of the model when both IASI and TROPOMI are assimilated.

#### 6 Impact of assimilation on forecasts

In this part, we study the impact of the assimilation on forecasts. To initialise the forecast, we use the assimilation outputs from the joint\_assim experiment. We use the term D0 for a 24 h range term forecast, D1 for a 48 h range term forecast and D2 for a 72 h range term forecast.

In Fig. 9, the fourth and the fifth line show the  $SO_2$  total column forecast for 12 April 2021 at 01:00, 13:00, 15:00, 16:00 and 17:00 UTC. These lines represent a forecast initialised by the 11 April 2021 analysis outputs and by the 10 April 2021 analysis outputs respectively. SO<sub>2</sub> total column observations are plotted on the first line for TROPOMI, the second and the third line for IASI, and the last line for OMI. When the model forecasts are initialised by the 9 April analysis outputs, no SO<sub>2</sub> total column value greater than 1 DU is present because even if the eruption started on 9 April, none of the assimilated instruments overpass the plume on this day. Using the results of the analysis on 10 April enables the model to simulate an SO<sub>2</sub> plume which reaches western Africa. The observations show a plume reaching not only Africa but also the vicinity of the volcano. The western part of the plume is not modelled by MOCAGE



**Figure 8.** Probability of detection (first line), critical success index (second line) and false alarm rate (last line) for the 1 and 5 DU thresholds for the three experiments: tropomi\_assim in blue, iasi\_assim in red and joint\_assim in green. The meaning of the symbols is described in Table 2.

on 12 April 2021 with the use of the 10 April analysis outputs because the last assimilation took place almost 2 d before. The more recent  $SO_2$  emissions can not be simulated. Nevertheless, the part of the already simulated plume matches well with the observed plume intensity. Using the latest available analysis outputs from 11 April, MOCAGE predicts a plume whose shape is closer to the observed one but is always smaller. However, the model tends to overestimate the total intensity of the  $SO_2$  column compared to the TROPOMI and IASI measurements. In addition, using the latest available analysis predicts an  $SO_2$  plume closer to the volcano.

Figure 10 represents the number of grid cells exceeding 1 and 5 DU for the D0 forecast in blue, for the D1 forecast in red and for the D2 forecast in green. The number of grid cells exceeding these thresholds is computed for TROPOMI in orange, IASI B in purple, IASI C in magenta and OMI in grey. We looked at the number of grid cells where the minimum and maximum of the total columns exceed these thresholds. These values are represented by horizontal lines in Fig. 10. The number of grid cells where the median of the observations exceeds these thresholds is shown as a dot. Finally, the limits of the bars represent the number of grid cells where the 25th and 75th quantiles exceed the thresholds.

Generally speaking, the number of meshes exceeding 1 or 5 DU decreases with the forecast period. This was not observed on 9 and 10 April, when no mesh exceeded 1 DU. The D0, D1 and D2 forecasts show the presence of a plume from 11, 12 and 13 April 2021. These forecasts are initialised by the output of the assimilation of 9 April, i.e. before the beginning of the eruption. On 11 April, there were around 200 grid cells where the model exceeded 1 DU for the D0 forecast. This number corresponds to the minimum number of grid cells where the TROPOMI and OMI observations reach 1 DU and is below the number of grid cells where the median of the IASI observations reaches 1 DU. On 12 April, the plume forecast with D0 was larger, with around 500 meshes exceeding 1 DU, corresponding to the number of meshes where the 25th quantile of the TROPOMI observations reached 1 DU and also where the median of the OMI observations reached this threshold. After this date, the number of occurrences of the SO<sub>2</sub> total column exceeding 1 DU increases and becomes greater than the number of grid cells where the IASI and OMI observations reach 1 DU. However, this number remains smaller than the maximum number of meshes calculated using TROPOMI observations. The number of points where the total column reaches 1 DU decreases



**Figure 9.** Observations and forecasts of  $SO_2$  total columns on 12 April 2021 at 01:00, 13:00, 15:00, 16:00 and 17:00 UTC. The first row and the last row correspond to TROPOMI and OMI observations respectively. Forecasts are computed from the 11 April analysis outputs (second row) and from the 10 April analysis outputs (third row). These forecasts are available for 12 April.

with the forecast term range. Nevertheless, this number always exceeds the number of grid cells where the OMI and IASI observations reach 1 DU from 14 April onwards. Regarding the 5 DU threshold, no grid cell exceeds this threshold for the D2 forecast. The D1 forecast shows a low number on 12 April when the total columns reach 5 DU. However, this number is similar to the number of grid cells where the median of TROPOMI and IASI observations reaches 5 DU. In addition, for this day, the number of points where the model reaches 5 DU is similar between the D0 and D1 forecasts. For 11 April, the number of occurrences of a total column greater than or equal to 5 DU is low in the model compared with the UV instruments. This number is within the range of grid cells where IASI observations exceed 5 DU. Figure 11 represents the POD (on the first line), the CSI (on the second line) and the FAR (on the third line) metrics calculated by comparing the observations measured by OMI and the forecasts at several time steps for 1 and 5 DU thresholds. The first forecast is launched on 9 April. Consequently, there is no D1 forecast available for this day and no D2 forecast available on 9 and 10 April. The last forecast is launched on 15 April, so there is no D1 forecast available on 16 and 17 April. There is no D1 forecast available on 17 April. For the POD and CSI metrics computed for the 1 DU threshold, the best values are found in the D0 forecast except on 14 and 15 April for the CSI metric. On 14 April CSI values are similar between the D0 and D1 forecast. Compared to POD



**Figure 10.** Number of grid cells where forecasts initialised by the joint\_assim outputs reach 1 and 5 DU. The blue, red and green lines show the number of points at which the total columns reach these thresholds in the D0, D1 and D2 forecasts. Orange, purple, magenta and grey boxplots represent the number of grid cells where TROPOMI, IASI B, IASI C and OMI observations exceed the threshold.

values, CSI values are much lower, especially with D0 forecasts on 13 and 14 April. It means that there are a lot of locations where MOCAGE wrongly simulated total columns greater than 1 DU. This finding is strengthened by the presence of the highest FAR values during these days. The POD and CSI metrics computed with a 5 DU threshold show similar values. These metrics are 0 except on 11 April, with values around 0.2 for both the POD and CSI metrics. On 12 April both misses and false alarms occur with the D0 and D1 forecasts. Elsewhere, no SO<sub>2</sub> total columns greater than 5 DU are modelled and no observations greater than this threshold are observed by OMI on 14, 15 and 17 April. With the 5 DU threshold, there are only correct rejections.

Incorporating  $SO_2$  total column analysis outputs into the MOCAGE forecasts enhances its ability to model an  $SO_2$  plume. In the absence of the assimilation, the model fails to forecast the presence of an  $SO_2$  plume. The use of recent analysis outputs progressively aligns the predicted location

of the plume with the observations. However, the predicted intensity of the plume remains subject to high uncertainty.

#### 7 Conclusion and perspectives

In this paper, we study the input of the assimilation of volcanic SO<sub>2</sub> total columns from TROPOMI, IASI, and both TROPOMI and IASI into the CTM MOCAGE in the case of the La Soufrière Saint Vincent eruption between 9 and 15 April 2021. For the background error covariance matrix, we used a profile containing high values in the volcanic plume vertical range extension. We considered a plume ranging from 9 to 21 km, corresponding to 90 % of the SO<sub>2</sub> plume heights diagnosed by the TROPOMI Layer Height product.

Thanks to the assimilation of TROPOMI and IASI data, an SO<sub>2</sub> plume is simulated in MOCAGE. This plume is modelled more or less early and corrected more or less often,



Figure 11. POD on the first line, CSI on the second line and FAR on the last line for the 1 and 5 DU thresholds for the D0 forecast in blue, the D1 forecast in red and the D2 forecast in green. The meaning of the symbols is described in Table 2.

depending on the time of the satellite overpass and on the instrument technology. The assimilation of both TROPOMI and IASI leads to a larger plume and a more realistic amount of  $SO_2$  in the model. During this eruption, MOCAGE is able to simulate the process of converting SO2 into sulfate aerosols. A sulfate plume, stronger by assimilating only TROPOMI, is computed by MOCAGE. With this creation of sulfate aerosols, AOD increases slightly. Nevertheless, few AOD observations are available during the studied period. Compared with SO<sub>2</sub> total columns observations, the number of pixels larger than 1 DU in the model is too large but the probability of detecting an SO<sub>2</sub> total column is important. In the model, the number of points with a total column greater than 5 DU is close to the number of grid cells with a median observation greater than 5 DU, especially when IASI and TROPOMI are assimilated. Compared to independent observations from OMI, the probability of detecting values greater than 5 DU is better when assimilating observations from both instruments.

Using assimilation outputs to compute forecasts improves the representation of  $SO_2$  total columns in the model. The size and the shape of the plume depends on the forecast range term. As the forecast time range increases, the size of the plume decreases. Sometimes and especially for low values of the  $SO_2$  total column, the size of the modelling plume is too large compared to the observations. Concerning the probability of detecting an  $SO_2$  total column greater than a threshold, it is generally better for the short-term forecast.

One potential source of discrepancy is the assumptions regarding the altitude and thickness of the plume, which are dynamic in space and time. An inaccurate representation of the  $SO_2$  plume height within the model could lead to a more dispersed plume or, depending on wind shear conditions, could result in the plume drifting in an incorrect direction. To refine our assimilation process, we could incorporate observed volcanic  $SO_2$  plume heights from both IASI and TROPOMI. This approach, as suggested by Inness et al. (2022), would likely yield a more accurate simulation of the altitude of the plume, thereby producing a modelled plume shape that better reflects reality.

Another factor contributing to the overestimation of the plume extent within the MOCAGE model is the assimilation settings. Specifically, the application of an excessively large horizontal and vertical correlation length could lead to too extended a plume both on the vertical and the horizontal dimensions. Moreover, the chosen standard deviation for the background error can significantly influence the simulated intensity of the plume. With too large a value, the SO<sub>2</sub> plume

concentration can be underestimated in MOCAGE, whereas with too small a value, the plume intensity can be too strong.

Additionally, inherent uncertainties in the model itself must be considered. The chemical processes involving  $SO_2$ , including reactions that may not be fully captured, introduce additional complexity into the simulation (Schumann et al., 2011). Furthermore, the meteorological data driving the model are not without their uncertainties (Webster and Thomson, 2022), which can compound the challenges in accurately modelling the transport and transformation of  $SO_2$  emissions.

Satellites can give information on the locations where no  $SO_2$  is detected. The use of this information for the assimilation could improve the process because it allows for limiting the shape of the plume.

To facilitate the assimilation of the SO<sub>2</sub>, particularly the convergence of the minimiser, we could also use a prior volcanic SO<sub>2</sub> emission. In fact, the model and the observations are very different, sometimes by several orders of magnitude. To estimate volcanic SO<sub>2</sub> emissions, a source inversion of the volcanic SO<sub>2</sub> could be used (Boichu et al., 2013).

We also have seen that the more frequently the model is corrected, the closer it is to the observations. Assimilating additional instruments would therefore improve assimilation of the SO<sub>2</sub>. The best option would be to assimilate observations from geostationary satellites covering the globe.

Finally, we made a specific adjustment for this eruption. Ideally, we would like to have a task running daily with assimilation settings that would allow us to assimilate the volcanic  $SO_2$  for each eruption.

*Code availability.* The code used to generate the analysis (MOCAGE and its variational assimilation suite) is researchoperational code that is property of Météo-France and CERFACS and is not publicly available yet. The readers interested in obtaining parts of the code for research purposes can contact the authors of this study directly.

*Data availability.* All results are available upon request to the authors for 5 years.

Author contributions. MB and VG designed this study. MB carried out and interpreted the simulations with JA during his internship supervised by MB and VG. MB is the main contributor to the manuscript. VG and JA reviewed and contributed to the manuscript.

*Competing interests.* The contact author has declared that none of the authors has any competing interests.

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#### References

- Bennis, K. and Venzke, E.: Report on Soufriere St. Vincent (Saint Vincent and the Grenadines), Bulletin of the Global Volcanism Network, 46, https://doi.org/10.5479/si.GVP.BGVN202105-360150, 2021.
- Boichu, M., Menut, L., Khvorostyanov, D., Clarisse, L., Clerbaux, C., Turquety, S., and Coheur, P.-F.: Inverting for volcanic SO<sub>2</sub> flux at high temporal resolution using spaceborne plume imagery and chemistry-transport modelling: the 2010 Eyjafjallajökull eruption case study, Atmos. Chem. Phys., 13, 8569–8584, https://doi.org/10.5194/acp-13-8569-2013, 2013.
- Bouyssel, F., Berre, L., Bénichou, H., Chambon, P., Girardot, N., Guidard, V., Loo, C., Mahfouf, J.-F., Moll, P., Payan, C., and Raspaud, D.: The 2020 Global Operational NWP Data Assimilation System at Météo-France, Springer International Publishing, 4, 645–664, https://doi.org/10.1007/978-3-030-77722-7\_25, 2022.
- Brenot, H., Theys, N., Clarisse, L., van Geffen, J., van Gent, J., Van Roozendael, M., van der A, R., Hurtmans, D., Coheur, P.-F., Clerbaux, C., Valks, P., Hedelt, P., Prata, F., Rasson, O., Sievers, K., and Zehner, C.: Support to Aviation Control Service (SACS): an online service for near-real-time satellite monitoring of volcanic plumes, Nat. Hazards Earth Syst. Sci., 14, 1099–1123, https://doi.org/10.5194/nhess-14-1099-2014, 2014.
- Brenot, H., Theys, N., Clarisse, L., van Gent, J., Hurtmans, D. R., Vandenbussche, S., Papagiannopoulos, N., Mona, L., Virtanen, T., Uppstu, A., Sofiev, M., Bugliaro, L., Vázquez-Navarro, M., Hedelt, P., Parks, M. M., Barsotti, S., Coltelli, M., Moreland, W., Scollo, S., Salerno, G., Arnold-Arias, D., Hirtl, M., Peltonen, T., Lahtinen, J., Sievers, K., Lipok, F., Rüfenacht, R., Haefele, A., Hervo, M., Wagenaar, S., Som de Cerff, W., de Laat, J., Apituley, A., Stammes, P., Laffineur, Q., Delcloo, A., Lennart, R., Rokitansky, C.-H., Vargas, A., Kerschbaum, M., Resch, C., Zopp, R., Plu, M., Peuch, V.-H., Van Roozendael, M., and Wotawa, G.: EUNADICS-AV early warning system dedicated to supporting aviation in the case of a crisis from natural airborne hazards and radionuclide clouds, Nat. Hazards Earth Syst. Sci., 21, 3367– 3405, https://doi.org/10.5194/nhess-21-3367-2021, 2021.
- Carn, S. A., Krueger, A. J., Krotkov, N. A., Yang, K., and Evans, K.: Tracking volcanic sulfur dioxide clouds for aviation hazard mitigation, Nat. Hazards, 51, 325–343, https://doi.org/10.1007/s11069-008-9228-4, 2009.

- Carn, S. A., Clarisse, L., and Prata, A. J.: Multidecadal satellite measurements of global volcanic degassing, J. Volcanol. Geoth. Res., 311, 99–134, https://doi.org/10.1016/j.jvolgeores.2016.01.002, 2016.
- Clarisse, L., Hurtmans, D., Clerbaux, C., Hadji-Lazaro, J., Ngadi, Y., and Coheur, P.-F.: Retrieval of sulphur dioxide from the infrared atmospheric sounding interferometer (IASI), Atmos. Meas. Tech., 5, 581–594, https://doi.org/10.5194/amt-5-581-2012, 2012.
- Clarisse, L., Coheur, P.-F., Theys, N., Hurtmans, D., and Clerbaux, C.: The 2011 Nabro eruption, a SO<sub>2</sub> plume height analysis using IASI measurements, Atmos. Chem. Phys., 14, 3095–3111, https://doi.org/10.5194/acp-14-3095-2014, 2014.
- Clerbaux, C., Boynard, A., Clarisse, L., George, M., Hadji-Lazaro, J., Herbin, H., Hurtmans, D., Pommier, M., Razavi, A., Turquety, S., Wespes, C., and Coheur, P.-F.: Monitoring of atmospheric composition using the thermal infrared IASI/MetOp sounder, Atmos. Chem. Phys., 9, 6041–6054, https://doi.org/10.5194/acp-9-6041-2009, 2009.
- Cornut, F., El Amraoui, L., Cuesta, J., Schmisser, R., Blanc, J., and Josse, B.: Assimilation of Aerosol Observations from the Future Spaceborne Lidar Onboard the AOS Mission into the MOCAGE Chemistry: Transport Model, in: Proceedings of the 30th International Laser Radar Conference, edited by: Sullivan, J. T., Leblanc, T., Tucker, S., Demoz, B., Eloranta, E., Hostetler, C., Ishii, S., Mona, L., Moshary, F., Papayannis, A., and Rupavatharam, K., Springer International Publishing, 645– 651, https://doi.org/10.1007/978-3-031-37818-8\_83, ISBN 978-3-031-37818-8, 2023.
- Courtier, P., Freydier, C., Geleyn, J.-F., and Rabier, F., and Rochas, M.: The Arpege project at Meteo France, ECMWF, II, 193–232, 1991.
- Cussac, M., Marécal, V., Thouret, V., Josse, B., and Sauvage, B.: The impact of biomass burning on upper tropospheric carbon monoxide: a study using MOCAGE global model and IAGOS airborne data, Atmos. Chem. Phys., 20, 9393–9417, https://doi.org/10.5194/acp-20-9393-2020, 2020.
- El Aabaribaoune, M., Emili, E., and Guidard, V.: Estimation of the error covariance matrix for IASI radiances and its impact on the assimilation of ozone in a chemistry transport model, Atmos. Meas. Tech., 14, 2841–2856, https://doi.org/10.5194/amt-14-2841-2021, 2021.
- El Amraoui, L., Sič, B., Piacentini, A., Marécal, V., Frebourg, N., and Attié, J.-L.: Aerosol data assimilation in the MOCAGE chemical transport model during the TRAQA/ChArMEx campaign: lidar observations, Atmos. Meas. Tech., 13, 4645–4667, https://doi.org/10.5194/amt-13-4645-2020, 2020.
- El Amraoui, L., Plu, M., Guidard, V., Cornut, F., and Bacles, M.: A Pre-Operational System Based on the Assimilation of MODIS Aerosol Optical Depth in the MOCAGE Chemical Transport Model, Remote Sens., 14, 1949, https://doi.org/10.3390/rs14081949, 2022.
- Emili, E., Barret, B., Massart, S., Le Flochmoen, E., Piacentini, A., El Amraoui, L., Pannekoucke, O., and Cariolle, D.: Combined assimilation of IASI and MLS observations to constrain tropospheric and stratospheric ozone in a global chemical transport model, Atmos. Chem. Phys., 14, 177–198, https://doi.org/10.5194/acp-14-177-2014, 2014.

- Esse, B., Burton, M., Hayer, C., Contreras-Arratia, R., Christopher, T., Joseph, E. P., Varnam, M., and Johnson, C.: SO<sub>2</sub> emissions during the 2021 eruption of La Soufrière, St Vincent, revealed with back-trajectory analysis of TROPOMI imagery, Geological Society of London, Special Publications, 539, 231–244, https://doi.org/10.1144/SP539-2022-77, 2024.
- Feinberg, A., Sukhodolov, T., Luo, B.-P., Rozanov, E., Winkel, L. H. E., Peter, T., and Stenke, A.: Improved tropospheric and stratospheric sulfur cycle in the aerosol–chemistry–climate model SOCOL-AERv2, Geosci. Model Dev., 12, 3863–3887, https://doi.org/10.5194/gmd-12-3863-2019, 2019.
- Flemming, J. and Inness, A.: Volcanic sulfur dioxide plume forecasts based on UV satellite retrievals for the 2011 Grímsvötn and the 2010 Eyjafjallajökull eruption, J. Geophys. Res.-Atmos., 118, 10172–10189, https://doi.org/10.1002/jgrd.50753, 2013.
- Gouhier, M., Deslandes, M., Guéhenneux, Y., Hereil, P., Cacault, P., and Josse, B.: Operational Response to Volcanic Ash Risks Using HOTVOLC Satellite-Based System and MOCAGE-Accident Model at the Toulouse VAAC, Atmosphere, 11, 864, https://doi.org/10.3390/atmos11080864, 2020.
- Guth, J., Josse, B., Marécal, V., Joly, M., and Hamer, P.: First implementation of secondary inorganic aerosols in the MOCAGE version R2.15.0 chemistry transport model, Geosci. Model Dev., 9, 137–160, https://doi.org/10.5194/gmd-9-137-2016, 2016.
- Hedelt, P., Efremenko, D. S., Loyola, D. G., Spurr, R., and Clarisse, L.: Sulfur dioxide layer height retrieval from Sentinel-5 Precursor/TROPOMI using FP\_ILM, Atmos. Meas. Tech., 12, 5503– 5517, https://doi.org/10.5194/amt-12-5503-2019, 2019.
- Huijnen, V., Williams, J., van Weele, M., van Noije, T., Krol, M., Dentener, F., Segers, A., Houweling, S., Peters, W., de Laat, J., Boersma, F., Bergamaschi, P., van Velthoven, P., Le Sager, P., Eskes, H., Alkemade, F., Scheele, R., Nédélec, P., and Pätz, H.-W.: The global chemistry transport model TM5: description and evaluation of the tropospheric chemistry version 3.0, Geosci. Model Dev., 3, 445–473, https://doi.org/10.5194/gmd-3-445-2010, 2010.
- Inness, A., Ades, M., Balis, D., Efremenko, D., Flemming, J., Hedelt, P., Koukouli, M.-E., Loyola, D., and Ribas, R.: Evaluating the assimilation of S5P/TROPOMI near real-time SO<sub>2</sub> columns and layer height data into the CAMS integrated forecasting system (CY47R1), based on a case study of the 2019 Raikoke eruption, Geosci. Model Dev., 15, 971–994, https://doi.org/10.5194/gmd-15-971-2022, 2022.
- Josse, B., Simon, P., and Peuch, V. H.: Radon global simulations with the multiscale chemistry and transport model MOCAGE, Tellus B, 56, 339–356, https://doi.org/10.3402/tellusb.v56i4.16448, 2004.
- Kaiser, J. W., Heil, A., Andreae, M. O., Benedetti, A., Chubarova, N., Jones, L., Morcrette, J.-J., Razinger, M., Schultz, M. G., Suttie, M., and van der Werf, G. R.: Biomass burning emissions estimated with a global fire assimilation system based on observed fire radiative power, Biogeosciences, 9, 527–554, https://doi.org/10.5194/bg-9-527-2012, 2012.
- Koukouli, M.-E., Michailidis, K., Hedelt, P., Taylor, I. A., Inness, A., Clarisse, L., Balis, D., Efremenko, D., Loyola, D., Grainger, R. G., and Retscher, C.: Volcanic SO<sub>2</sub> layer height by TROPOMI/S5P: evaluation against IASI/MetOp and CALIOP/-CALIPSO observations, Atmos. Chem. Phys., 22, 5665–5683, https://doi.org/10.5194/acp-22-5665-2022, 2022.

- Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., Van Aardenne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V., Riahi, K., and van Vuuren, D. P.: Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application, Atmos. Chem. Phys., 10, 7017–7039, https://doi.org/10.5194/acp-10-7017-2010, 2010.
- Lamotte, C., Guth, J., Marécal, V., Cussac, M., Hamer, P. D., Theys, N., and Schneider, P.: Modeling study of the impact of SO<sub>2</sub> volcanic passive emissions on the tropospheric sulfur budget, Atmos. Chem. Phys., 21, 11379–11404, https://doi.org/10.5194/acp-21-11379-2021, 2021.
- Lechner, P., Tupper, A., Guffanti, M., Loughlin, S., and Casadevall, T.: Volcanic Ash and Aviation – The Challenges of Real-Time, Global Communication of a Natural Hazard, in: Observing the Volcano World: Volcano Crisis Communication, edited by: Fearnley, C. J., Bird, D. K., Haynes, K., McGuire, W. J., and Jolly, G., 51–64, https://doi.org/10.1007/11157\_2016\_49, ISBN 978-3-319-44097-2, 2018.
- Lefevre, F., Brasseur, G., Folkins, I., Smith, A., and Simon, P.: Chemistry of the 1991–1992 stratospheric winter: Threedimensional model simulations, J. Geophys. Res.-Atmos., 99, 8183–8195, https://doi.org/10.1029/93JD03476, 1994.
- Li, C., Krotkov, N. A., Carn, S., Zhang, Y., Spurr, R. J. D., and Joiner, J.: New-generation NASA Aura Ozone Monitoring Instrument (OMI) volcanic SO<sub>2</sub> dataset: algorithm description, initial results, and continuation with the Suomi-NPP Ozone Mapping and Profiler Suite (OMPS), Atmos. Meas. Tech., 10, 445– 458, https://doi.org/10.5194/amt-10-445-2017, 2017.
- Marécal, V., Peuch, V.-H., Andersson, C., Andersson, S., Arteta, J., Beekmann, M., Benedictow, A., Bergström, R., Bessagnet, B., Cansado, A., Chéroux, F., Colette, A., Coman, A., Curier, R. L., Denier van der Gon, H. A. C., Drouin, A., Elbern, H., Emili, E., Engelen, R. J., Eskes, H. J., Foret, G., Friese, E., Gauss, M., Giannaros, C., Guth, J., Joly, M., Jaumouillé, E., Josse, B., Kadygrov, N., Kaiser, J. W., Krajsek, K., Kuenen, J., Kumar, U., Liora, N., Lopez, E., Malherbe, L., Martinez, I., Melas, D., Meleux, F., Menut, L., Moinat, P., Morales, T., Parmentier, J., Piacentini, A., Plu, M., Poupkou, A., Queguiner, S., Robertson, L., Rouïl, L., Schaap, M., Segers, A., Sofiev, M., Tarasson, L., Thomas, M., Timmermans, R., Valdebenito, Á., van Velthoven, P., van Versendaal, R., Vira, J., and Ung, A.: A regional air quality forecasting system over Europe: the MACC-II daily ensemble production, Geosci. Model Dev., 8, 2777-2813, https://doi.org/10.5194/gmd-8-2777-2015, 2015.
- Prata, A. J.: Satellite detection of hazardous volcanic clouds and the risk to global air traffic, Nat. Hazards, 51, 303–324, https://doi.org/10.1007/s11069-008-9273-z, 2009.
- Prata, F., Woodhouse, M., Huppert, H. E., Prata, A., Thordarson, T., and Carn, S.: Atmospheric processes affecting the separation of volcanic ash and SO<sub>2</sub> in volcanic eruptions: inferences from the May 2011 Grímsvötn eruption, Atmos. Chem. Phys., 17, 10709– 10732, https://doi.org/10.5194/acp-17-10709-2017, 2017.
- Price, C., Penner, J., and Prather, M.: NO<sub>x</sub> from lightning: 1. Global distribution based on lightning physics, J. Geophys. Res.-Atmos., 102, 5929–5941, https://doi.org/10.1029/96JD03504, 1997.

- Qu, Z., Henze, D. K., Li, C., Theys, N., Wang, Y., Wang, J., Wang, W., Han, J., Shim, C., Dickerson, R. R., and Ren, X.: SO<sub>2</sub> Emission Estimates Using OMI SO<sub>2</sub> Retrievals for 2005–2017, J. Geophys. Res.-Atmos., 124, 8336–8359, https://doi.org/10.1029/2019JD030243, 2019.
- Reshi, A. R., Pichuka, S., and Tripathi, A.: Applications of sentinel-5p tropomi satellite sensor: A review, IEEE Sens. J., 24, 20312– 20321, 2024.
- Rodgers, C. D. and Connor, B. J.: Intercomparison of remote sounding instruments, J. Geophys. Res.-Atmos., 108, 4116, https://doi.org/10.1029/2002JD002299, 2003.
- Rouil, L., Honoré, C., Vautard, R., Beekmann, M., Bessagnet, B., Malherbe, L., Meleux, F., Dufour, A., Elichegaray, C., Flaud, J.-M., Menut, L., Martin, D., Peuch, A., Peuch, V.-H., and Poisson, N.: Prev'air: An Operational Forecasting and Mapping System for Air Quality in Europe, B. Am. Meteorol. Soc., 90, 73–84, https://doi.org/10.1175/2008BAMS2390.1, 2009.
- Schmidt, A., Witham, C. S., Theys, N., Richards, N. A. D., Thordarson, T., Szpek, K., Feng, W., Hort, M. C., Woolley, A. M., Jones, A. R., Redington, A. L., Johnson, B. T., Hayward, C. L., and Carslaw, K. S.: Assessing hazards to aviation from sulfur dioxide emitted by explosive Icelandic eruptions, J. Geophys. Res.-Atmos., 119, 14180–14196, https://doi.org/10.1002/2014JD022070, 2014.
- Schumann, U., Weinzierl, B., Reitebuch, O., Schlager, H., Minikin, A., Forster, C., Baumann, R., Sailer, T., Graf, K., Mannstein, H., Voigt, C., Rahm, S., Simmet, R., Scheibe, M., Lichtenstern, M., Stock, P., Rüba, H., Schäuble, D., Tafferner, A., Rautenhaus, M., Gerz, T., Ziereis, H., Krautstrunk, M., Mallaun, C., Gayet, J.-F., Lieke, K., Kandler, K., Ebert, M., Weinbruch, S., Stohl, A., Gasteiger, J., Groß, S., Freudenthaler, V., Wiegner, M., Ansmann, A., Tesche, M., Olafsson, H., and Sturm, K.: Airborne observations of the Eyjafjalla volcano ash cloud over Europe during air space closure in April and May 2010, Atmos. Chem. Phys., 11, 2245–2279, https://doi.org/10.5194/acp-11-2245-2011, 2011.
- Sears, T. M., Thomas, G. E., Carboni, E., A. Smith, A. J., and Grainger, R. G.: SO<sub>2</sub> as a possible proxy for volcanic ash in aviation hazard avoidance, J. Geophys. Res.-Atmos., 5698–5709, https://doi.org/10.1002/jgrd.50505, 2013.
- Sellitto, P., Podglajen, A., Belhadji, R., Boichu, M., Carboni, E., Cuesta, J., Duchamp, C., Kloss, C., Siddans, R., BÃ<sup>-</sup>gue, N., Blarel, L., Jegou, F., Khaykin, S., Renard, J.-B., and Legras, B.: The unexpected radiative impact of the Hunga Tonga eruption of 15th January 2022, Communications Earth & Environment, 3, 1–10, https://doi.org/10.1038/s43247-022-00618-z, 2022.
- Sindelarova, K., Granier, C., Bouarar, I., Guenther, A., Tilmes, S., Stavrakou, T., Müller, J.-F., Kuhn, U., Stefani, P., and Knorr, W.: Global data set of biogenic VOC emissions calculated by the MEGAN model over the last 30 years, Atmos. Chem. Phys., 14, 9317–9341, https://doi.org/10.5194/acp-14-9317-2014, 2014.
- Sič, B., El Amraoui, L., Marécal, V., Josse, B., Arteta, J., Guth, J., Joly, M., and Hamer, P. D.: Modelling of primary aerosols in the chemical transport model MOCAGE: development and evaluation of aerosol physical parameterizations, Geosci. Model Dev., 8, 381–408, https://doi.org/10.5194/gmd-8-381-2015, 2015.
- Sič, B., El Amraoui, L., Piacentini, A., Marécal, V., Emili, E., Cariolle, D., Prather, M., and Attié, J.-L.: Aerosol data assimilation in the chemical transport model MOCAGE during the TRAQA/ChArMEx campaign: aerosol optical depth, Atmos.

Meas. Tech., 9, 5535–5554, https://doi.org/10.5194/amt-9-5535-2016, 2016.

- Spurr, R., de Haan, J., van Oss, R., and Vasilkov, A.: Discreteordinate radiative transfer in a stratified medium with first-order rotational Raman scattering, J. Quant. Spectrosc. Ra., 109, 404– 425, https://doi.org/10.1016/j.jqsrt.2007.08.011, 2008.
- Stockwell, W. R., Kirchner, F., Kuhn, M., and Seefeld, S.: A new mechanism for regional atmospheric chemistry modeling, J. Geophys. Res.-Atmos., 102, 25847–25879, https://doi.org/10.1029/97JD00849, 1997.
- Theys, N.: S5P/TROPOMI SO<sub>2</sub> ATBD, Tech. rep. 1.1.0, BIRA, https://sentinels.copernicus.eu/documents/247904/2476257/ Sentinel-5P-ATBD-SO2-TROPOMI/c3930dc9-16f6-4403-9049 -db768eca6ff9?version=1.1 (last access: 17 June 2025), 2018.
- Theys, N., De Smedt, I., Yu, H., Danckaert, T., van Gent, J., Hörmann, C., Wagner, T., Hedelt, P., Bauer, H., Romahn, F., Pedergnana, M., Loyola, D., and Van Roozendael, M.: Sulfur dioxide retrievals from TROPOMI onboard Sentinel-5 Precursor: algorithm theoretical basis, Atmos. Meas. Tech., 10, 119–153, https://doi.org/10.5194/amt-10-119-2017, 2017.
- Theys, N., Hedelt, P., De Smedt, I., Lerot, C., Yu, H., Vlietinck, J., Pedergnana, M., Arellano, S., Galle, B., Fernandez, D., Carlito, C. J. M., Barrington, C., Taisne, B., Delgado-Granados, H., Loyola, D., and Van Roozendael, M.: Global monitoring of volcanic SO<sub>2</sub> degassing with unprecedented resolution from TROPOMI onboard Sentinel-5 Precursor, Scientific Reports, 9, 2643, https://doi.org/10.1038/s41598-019-39279-y, 2019.

- Thomas, H. E. and Prata, A. J.: Sulphur dioxide as a volcanic ash proxy during the April–May 2010 eruption of Eyjafjallajökull Volcano, Iceland, Atmos. Chem. Phys., 11, 6871–6880, https://doi.org/10.5194/acp-11-6871-2011, 2011.
- Veefkind, J. P., Aben, I., McMullan, K., Förster, H., de Vries, J., Otter, G., Claas, J., Eskes, H. J., de Haan, J. F., Kleipool, Q., van Weele, M., Hasekamp, O., Hoogeveen, R., Landgraf, J., Snel, R., Tol, P., Ingmann, P., Voors, R., Kruizinga, B., Vink, R., Visser, H., and Levelt, P. F.: TROPOMI on the ESA Sentinel-5 Precursor: A GMES mission for global observations of the atmospheric composition for climate, air quality and ozone layer applications, Remote Sens. Environ., 120, 70–83, https://doi.org/10.1016/j.rse.2011.09.027, 2012.
- Vittorioso, F., Guidard, V., and Fourrié, N.: Assessment of the contribution of the Meteosat Third Generation Infrared Sounder (MTG-IRS) for the characterisation of ozone over Europe, Atmos. Meas. Tech., 17, 5279–5299, https://doi.org/10.5194/amt-17-5279-2024, 2024.
- Webster, H. N. and Thomson, D. J.: Using ensemble meteorological data sets to treat meteorological uncertainties in a Bayesian volcanic ash inverse modeling system: A case study, Grímsvötn 2011, J. Geophys. Res.-Atmos., 127, e2022JD036469, https://doi.org/10.1029/2022JD036469, 2022.
- Witze, A.: Why the Tongan eruption will go down in the history of volcanology, Nature, 602, 376–378, https://doi.org/10.1038/d41586-022-00394-y, 2022.