# RESEARCH

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# A modelling approach for quantifying volcanic sulphur dioxide concentrations at flight altitudes and the potential hazard to aircraft occupants

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# Abstract

Volcanic eruptions can emit large quantities of sulphur dioxide (SO<sub>2</sub>) into the atmosphere, which can be harmful to people and the environment. Aircraft encounters with a volcanic SO<sub>2</sub> cloud could represent a health hazard to crew and passengers onboard. In this study we have assessed concentration levels of volcanic SO<sub>2</sub> in the atmosphere following eight historic eruptions and use four-dimensional dispersion model simulation data to calculate when and where the World Health Organisation (WHO) health protection guideline for SO<sub>2</sub> of 500  $\mu$ gm<sup>-3</sup> over 10 minutes is exceeded. The time and area of exceedance varies and depends on the eruption characteristics: the amount, duration and height of the SO<sub>2</sub> release. The WHO-based guideline value is exceeded for all historic eruptions considerably larger than the volcanic ash hazard area for the same eruption. SO<sub>2</sub> hazard areas also often extend over a longer period of time compared to the equivalent ash advisories. For example, following the 2019 eruption of Raikoke, the SO<sub>2</sub> hazard area reached up to 1.7 million km<sup>2</sup> and the WHO-based guideline value was exceeded for about two weeks, while volcanic ash was considered hazardous to aviation for about five days. These results will help the aviation industry to better understand the potential risks to their passengers and crew from volcanic SO<sub>2</sub>, and aid in defining concentration thresholds for any potential volcanic SO<sub>2</sub> forecasts for aviation.

**Keywords** Volcanic eruptions, Volcanic gases, Volcanic hazard, Sulphur dioxide, Hazard assessment, Exposure thresholds, Atmospheric models, Kasatochi, Raikoke

# Introduction

Volcanic activity can release significant quantities of gas, including sulphur dioxide  $(SO_2)$ , into our atmosphere (Bluth et al. 1993). Volcanic  $SO_2$  emissions can have important impacts on atmospheric chemistry and climate, terrestrial and marine environments, and human and animal health (e.g., Oppenheimer et al. 2011; Mather

\*Correspondence: N. I. Kristiansen nina.kristiansen@metoffice.gov.uk <sup>1</sup> Met Office, Exeter EX1 3PB, UK 2015). In this paper, we focus on the hazard posed to human health:  $SO_2$  is a fast-acting respiratory irritant when inhaled and at high concentrations may cause breathing difficulties (Orellano, Reynoso & Quaranta 2021). Those suffering from asthma and chronic lung disease may be especially susceptible to the adverse effects of  $SO_2$  and, exposure to high concentrations may provoke attacks of asthma. Based on the findings from experimental studies with exercising asthmatics, the World Health Organization (WHO) has defined an air quality guideline for short term  $SO_2$  exposure of 500 µgm<sup>-3</sup> over a 10-minute averaging time (WHO 2005, 2021). Due to



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its health impacts,  $SO_2$  is part of the air pollution forecast in many countries to mitigate the hazard from anthropogenic pollution sources (e.g., DAQI, 2022).

 $SO_2$  is a colourless toxic gas with a characteristic odour. Reported odour detection thresholds of  $SO_2$  (i.e., the threshold for noticing smell) range from 266 µgm<sup>-3</sup> to 12500 µgm<sup>-3</sup>, with median thresholds between 1766 µgm<sup>-3</sup> and 3575 µgm<sup>-3</sup> depending on age and gender (Kleinbeck et al. 2011). This implies that at the WHO guideline of 500 µgm<sup>-3</sup> most humans are not likely to smell  $SO_2$ and will only start smelling it once concentrations reach around a factor between 3 and 7 over the WHO guideline value. Consequently, harmful effects may be possible to susceptible individuals before they are aware of their exposure.

Each year several volcanic eruptions release significant amounts of SO<sub>2</sub> to the atmosphere. Carn et al. (2016) show that between 2008 and 2014 there were between 1 and 3 eruptions annually that released more than 100 kilotons (kt) of volcanic SO<sub>2</sub>. Volcanic emissions of SO<sub>2</sub> occur over a range of scales through different types of volcanic activity. Persistently degassing volcanoes like Kilauea (Hawaii), passively release gas at the surface and represent a significant source of gases to the global atmospheric budget (Carn et al. 2017). On average, over the past decade, a total of around 60 kt per day of  $SO_2$  has been released due to passive degassing from the world's volcanoes (Carn et al. 2017). Effusive eruptions release larger quantities of gas into the troposphere. For example, the 6-month Holuhraun eruption in Iceland during 2014, released around 60-100 kt/day of  $SO_2$  with a total of about 11000 kt, which is more than the anthropogenic amount emitted from Europe in 2011 (Gíslason et al. 2015). Large explosive eruptions can inject vast quantities of  $SO_2$  over a short period of time, and high into the troposphere and stratosphere where it can remain for days to weeks. The largest explosive eruption in the last 100 years was the 1991 eruption of Mt Pinatubo, which released 20000 kt of  $SO_2$  up to an altitude of more than 30 km (McCormick et al. 1995) and the SO<sub>2</sub> cloud was then rapidly advected around the globe. In the last 20 years, the largest explosive emitters of SO<sub>2</sub> were Kasatochi (~1700 kt) in 2008, Nabro (~3500 kt) in 2011 and Raikoke (~1500 kt) in 2019 (Carn et al. 2016, 2021). The volcanic SO<sub>2</sub> clouds originating from these eruptions were detected and tracked by several satellite instruments over a period of several weeks (e.g., Theys et al. 2013; de Leeuw et al. 2021).

Some eruptions release  $SO_2$  as well as volcanic ash, while some release only one or the other. If both are emitted then, depending on the eruption dynamics and meteorological conditions, they can be co-located and transported together or be separated and travel in different directions as separate clouds (e.g., Schneider et al. 1999; Prata et al. 2017).  $SO_2$  is often detected and transported over larger distances compared with volcanic ash (e.g., Prata et al. 2010). Removal of  $SO_2$  from the atmosphere happens by chemical reactions, dry and wet deposition, and uptake on atmospheric aerosols including volcanic ash (Martin et al. 2014, 2018; Zhu et al. 2020). The atmospheric lifetime of  $SO_2$  varies from a few days to several weeks and generally increases with altitude (Carn et al. 2016). The potential for long range transport of volcanic  $SO_2$  clouds consequently means they are likely to intersect global air traffic routes.

Between 1980 and 2016 there were at least 85 incidents where commercial aircraft crew reported encounters with volcanic SO<sub>2</sub>, with many of the reports identifying sulphurous smells in the cabin (Pers. Comm. D. J. Schneider, January 2024, unpublished observations; ICAO, 2015; Guffanti, Casadevall, and Budding, 2010b). When volcanic SO<sub>2</sub> enters an aircraft, its odour can cause distress to passengers and crew, and it may represent a significant health hazard if it is present at high enough concentrations and durations. In August 2008, after the eruption of the Kasatochi volcano in Alaska, several pilot reports specified aircraft crew smelling sulphur (Guffanti et al. 2010a, b). The reports were from flights along the west coast of Canada and northern USA where the SO<sub>2</sub> cloud from the eruption was located after it had been transported eastwards from the volcano. After the Grímsvötn 2011 eruption in Iceland, pilots reported unpleasant smells of sulphur while flying across the North Atlantic (European Space Agency 2012). It should be noted that to date, no health consequences from aircraft encounters with volcanic SO<sub>2</sub> clouds have been reported in the literature.

As these reports are from commercial aircraft, no measurements of the encountered SO<sub>2</sub> concentrations are available. However, measurements exist for two Icelandic volcanic  $SO_2$  clouds that were sampled by research aircraft. Following the Hekla 2000 eruption, in-situ measurements of cloud properties were measured by a research aircraft one and a half days after the eruption (Rose et al. 2003). The aircraft flew in the  $SO_2$  cloud for 7-10 minutes, during which time the on-board instrumentation recorded concentrations up to 1 ppm (about six times the WHO guideline, see further details on unit conversion later). The volcanic  $SO_2$  cloud from the 2010 Eyjafjallajökull eruption was also sampled by multiple research aircraft at various locations over Europe and measured  $SO_2$  values were up to 0.1 ppm (about half the WHO guideline) (Johnson et al. 2012; Schumann et al. 2011).

Schmidt et al. (2014) discuss the hazards to aviation from  $SO_2$  emitted by explosive Icelandic eruptions and

use model simulations to estimate  $SO_2$  concentrations that could occur in European and North Atlantic airspace following real and hypothetical eruptions in Iceland. They concluded that the WHO guideline was not exceeded in the far field ( $\geq$ 1000 km) for the 2010 Eyjafjallajökull eruption, and that there was a low risk of plume encounters exceeding the WHO guideline for a larger Hekla 2000 type eruption.

It is the responsibility of 9 worldwide Volcanic Ash Advisory Centres (VAACs) to provide operational forecasts of the expected transport and dispersion of volcanic ash clouds (ICAO, 2012), but there are currently no requirements to provide advice to aviation on the presence of volcanic SO<sub>2</sub> clouds. There are also no criteria which define when airspace is considered hazardous due to the presence of volcanic SO<sub>2</sub> clouds. However, the International Civil Aviation Organization (ICAO) is currently considering the introduction of global SO<sub>2</sub> forecast capability to provide advice to the aviation industry, with a focus on identifying and quantifying health risks to aircraft occupants (ICAO, 2018). This includes an assessment of the forecasting requirements for volcanic SO<sub>2</sub>.

The aim of this paper is to illustrate the implications of applying the WHO  $SO_2$  air quality guideline to determine potentially hazardous areas for aviation due to volcanic  $SO_2$  clouds. This is achieved by analysing modelled volcanic  $SO_2$  clouds from several historic eruptions that span a range of activity and emission scales.

The paper is organised as follows. In Section "Methods" we discuss the analysis methods including the dispersion model simulations conducted and the eruption cases considered. The results are presented in Section "Results" with a detailed focus on the 2008 Kasatochi eruption. In Section "Discussion" we discuss some of the limitations and uncertainties in our results, and a summary and conclusions are given in Section "Conclusions".

# Methods

Atmospheric dispersion model data for historic eruptions have been used to calculate the area and time over which  $SO_2$  concentration thresholds are exceeded. Here we first describe the numerical model used, followed by the method to calculate the exceedance area and duration based on  $SO_2$  concentration thresholds.

The Numerical Atmospheric-dispersion Modelling Environment (NAME) (Jones et al. 2007) is an atmospheric dispersion model capable of simulating many atmospheric dispersion phenomena and associated physical and chemical processes. These include emissions from nuclear accidents, volcanic eruptions, chemical accidents, smoke from fires, odours and airborne animal diseases, amongst others.

NAME is the operational model used at the London VAAC for forecasting volcanic ash (Beckett et al. 2020) and has also been used for simulating volcanic SO<sub>2</sub> clouds (Heard et al. 2012; Schmidt et al. 2014, 2015) including most recently a detailed analysis of the 2019 eruption of Raikoke (de Leeuw et al. 2021; Osborne et al. 2022). These previous studies have shown that NAME is able to capture the atmospheric transport and removal of volcanic  $SO_2$  clouds, providing that appropriate  $SO_2$ emission fluxes and meteorological data are used to drive the model. In particular, the study by de Leeuw et al (2021) demonstrated that for the 2019 Raikoke eruption, NAME was able to forecast the horizontal extent of the SO<sub>2</sub> cloud for 12-17 days after the initial eruption, and smaller scale features within the  $SO_2$  cloud with a skill on the order of 2-4 days.

NAME includes a chemistry scheme that converts  $SO_2$ into sulphur aerosols via reaction with climatological oxidants. The scheme includes both aqueous and gaseous phase chemistry reactions (Redington et al. 2009). Sulphate aerosol is produced in the gas phase by reaction with the hydroxyl radical (OH) to form  $SO_3$  which is then instantaneously converted into particulate sulphate. More details on the chemistry scheme can be found in de Leeuw et al. (2021).  $SO_2$  is also removed from the model atmosphere by wet and dry deposition (Webster and Thomson 2011, 2014).

NAME is an offline model and requires numerical weather prediction data (e.g., wind field data) for the simulation of advection and other atmospheric processes such as removal by precipitation. In our study, the NAME simulations were driven with the Met Office's global Unified Model (MetUM) meteorological data. The horizon-tal resolution of the meteorological data varies between around 40 km and 10 km depending on the year of the eruption. For the 2008-Kasatochi simulation the resolution is 0.5625° longitude by 0.375° latitude (~40 km), while for the 2019-Raikoke simulation the resolution is about 10 km.

The NAME output produced in this study was gridded fields with 30 min temporal resolution. The horizontal resolution of the gridded output matches the resolution of the historical meteorological data used to drive each simulation. The vertical resolution of the output is 500 m up to 20 km above sea level (asl) and 1 km resolution from 20 km to 23 km asl, giving a total of 43 altitude levels. Model output of atmospheric column loading values in Dobson Units (DU) is also used in this study, which means that we can directly compare and validate the model data to satellite observation. Quantitative information on atmospheric SO<sub>2</sub> retrieved from nadir sounding satellite instruments is often expressed as a column

loading in DU, where 1 DU equals 2.69 x  $10^{16}$  SO<sub>2</sub> molecules per cm<sup>2</sup>.

NAME is a hybrid Lagrangian-Eulerian model. In this work we only use the Lagrangian part which moves particles around in the modelled atmosphere. The chemistry is performed on the concentration field that is calculated from the Lagrangian particles.

Here we have used NAME to simulate the atmospheric transport and removal of volcanic  $SO_2$  from eight different volcanic eruptions (Table 1). The eruptions considered span a large range of eruption styles and sizes, with differences in the amount of  $SO_2$  released, the release height and duration. The evaluation presented is intended as a broad overview of types of events that could impact air traffic.

To initiate an  $SO_2$  model simulation with NAME, an appropriate estimate of the SO<sub>2</sub> emission flux and release height is needed, referred to as the  $SO_2$  source term. The source terms used in the NAME simulations are based on values reported in the literature. All source terms used have been derived by satellite data, either alone or in combination with modelling (e.g., inverse modelling). For Holuhraun, the SO<sub>2</sub> emission estimate from Schmidt et al. (2015) was used, releasing 60-100 kt/day uniformly between 1.5 and 3.0 km above ground level. We have considered only the first 2 months of the eruption when the largest SO<sub>2</sub> emissions occurred. For the Puyehue Cordon Caulle eruption, the source term determined from inverse modelling by Theys et al. (2013) was used with a uniform vertical distribution of the SO<sub>2</sub> release between 11-14 km asl. For Kasatochi, the inversion-based source term by Kristiansen et al. (2010) was applied, and for Raikoke, the source term estimated by de Leeuw et al. (2021) was used. Both eruptions were modelled with a non-uniform vertical distribution where more SO<sub>2</sub> was released at higher altitudes; 60-70% of the total  $SO_2$  emissions were released above the tropopause. For the four other eruptions (Fogo, Merapi, Kliuchevskoi and Etna) the source terms were taken from daily emissions and plume altitudes of SO<sub>2</sub> reported in a long-term database of volcanic SO<sub>2</sub> emissions derived from ultraviolet satellite measurements (Carn et al., 2021). The vertical extent of the release for these four eruptions was based on the observed plume altitude reported in the database. The Fogo SO<sub>2</sub> emissions were distributed uniformly from the summit height of the volcano, up to 7-9 km asl (the reported plume altitude). Merapi emissions were distributed uniformly between 10-17 km asl, Kliuchevskoi between 6-10 km asl, and Etna between 3.5-4.5 km asl. Tables including source term details for each eruption are provided in the Supplementary material. Uncertainties in the source emissions, the meteorological data and the modelled processes are discussed in Section "Discussion".

Using the model results, concentration thresholds for  $SO_2$  are applied to calculate an exceedance area and time. Table 2 lists the health relevant concentrations limits used in this analysis, which are informed by WHO guidelines and  $SO_2$  odour threshold values. In addition to the WHO short-term exposure (10 min) guideline, additional higher  $SO_2$  concentration thresholds were included to evaluate the impact on our results of using thresholds

**Table 2** Health relevant  $SO_2$  concentration limits considered in this analysis, informed by WHO guidelines and  $SO_2$  odour threshold values

Descriptor	Mass concentration at ground level (µgm <sup>-3</sup> )	Volumetric concentration (ppm)	
WHO	500	0.175	
WHOx2	1000	0.350	
WHOx6	3000	1.050	

**Table 1** List of eruptions analysed in this study. The emission height gives the vertical range where the largest emissions occur. Note that for the Holuhraun eruption (\*) the emissions were defined as above ground level, and the present analysis covers the first two months of the eruption period

_	Volcano	Eruption type	Country	Eruption start date	Eruption duration	Emission height (km asl)	Total SO <sub>2</sub> emission (kt)
1	Kliuchevskoi	Explosive	Russia	18/10/2013	8 days	6 – 10	55
2	Etna	Explosive	Italy	30/12/2013	1 day	3.5 - 4.5	10
3	Fogo	Effusive	Cape Verde	24/11/2014	34 days	3 - 9	382
4	Holuhraun	Effusive	Iceland	31/08/2014	~6 months*	~2 - 3*	~11000
5	Merapi	Explosive	Indonesia	04/11/2010	5 days	10 - 17	350
6	Puyehue	Explosive	Chile	04/06/2011	41 hours	11 - 14	155
7	Raikoke	Explosive	Russia	21/06/2019	9 hours	10 - 14	1570
8	Kasatochi	Explosive	Alaska	07/08/2008	< 1 hour	7 - 13	1700

that are closer to the odour threshold for  $SO_2$  (about 3-7 times above the WHO-based guideline). Multiples of the WHO-based guideline were chosen (WHOx2 and WHOx6) to evaluate whether this leads to a linear reduction in exceedance area and duration.

In order to apply the thresholds equally at all altitudes in the atmosphere, we first need to convert mass concentration thresholds (i.e., a threshold for the mass of a chemical per volume of air such as the WHO guideline of 500  $\mu$ gm<sup>-3</sup>) to volumetric units in parts per million (ppm). The amount of gas represented by a fixed mass concentration varies with atmospheric pressure and air density and therefore with altitude. Hence, it is not appropriate to apply one mass concentration threshold throughout the atmosphere for this type of analysis. Instead, we use a volumetric unit that is independent of altitude. The WHO guideline of 500 µgm<sup>-3</sup>, which is specific to ground level, equates to a volumetric unit of 0.175 ppm when using a conversion factor based on the molecular weight of SO<sub>2</sub> (64.06 gmol<sup>-1</sup>) and a "typical" surface atmospheric temperature and pressure of 20 degrees Celsius and 1013 millibar respectively. In this analysis we use 0.175 ppm as the WHO-based guideline at all altitude levels in the atmosphere. The same unit conversion is carried out for the additional higher thresholds.

We use the term 'hazard area' for the horizontal extent of the SO<sub>2</sub> cloud that exceeds a given concentration threshold (in unit of ppm). This is calculated from the 3-dimensional (latitude, longitude, altitude) model output with the following approach: if an SO<sub>2</sub> threshold is exceeded at any altitude level for a given horizonal grid box (longitude, latitude), the horizontal area of the grid box is added to the hazard area. This is subsequently repeated for all horizontal grid boxes in the modelled domain. This approach is used to provide a simplified 2-D assessment based on what in reality is a complex 3-D problem. This 2-D assessment also allows us to compare the SO<sub>2</sub> hazard area to 2-D volcanic ash location data from the Volcanic Ash Advisories (VAAs), issued by the responsible VAAC at the time of the eruption.

# Results

The NAME simulated SO<sub>2</sub> transport for the two largest eruptions considered in this analysis, the 2019 Raikoke and 2008 Kasatochi events, is shown in Fig. 1. These two eruptions were similar in terms of their emissions, with comparable total amounts of SO<sub>2</sub> and the same peak emission altitude of ~12 km asl. Satellite observations showed that the initial transport of the volcanic SO<sub>2</sub> clouds was similar as both were swept into passing low-pressure systems and showed distinct circular shaped clouds about 48 hours after the eruption onset (de Leeuw et al. 2021; Kristiansen et al. 2010). However, the further transport patterns differed significantly between these two eruptions (Fig. 1) due to differences in the meteorological conditions. The Kasatochi cloud was transported mainly eastwards, spreading and dispersing quickly, reaching Europe after about a week (Kristiansen et al. 2010), while the main part of the Raikoke cloud circulated over the North Pacific as a more compact entity for the first two weeks (de Leeuw et al. 2021). Detailed model validations of these two events can be found in de Leeuw et al. (2021), Heard et al. (2012) and Kristiansen et al. (2010), showing that dispersion models, including NAME, were able to simulate the volcanic SO<sub>2</sub> clouds with high accuracy over several days to weeks.

We now take a closer look at the Kasatochi SO<sub>2</sub> cloud for the time when several pilot reports along the Canadian west coast indicated sulphur smells in the cabin. Figure 2 shows the modelled SO<sub>2</sub> cloud at 21:30 UTC on 10 August 2008, 3 days after the eruption onset when the  $SO_2$  cloud had reached the Canadian west coast (Fig. 2a). One aircraft encounter reported sulphur odours at around 01:00 UTC on 11 August (Guffanti et al. 2010a, b). The model output is shown for 21:30 UTC to align with the VAA polygon (Fig. 2d) as no VAA output is available at the time of the encounter at 01:00 UTC. The approximate location of the encounter is marked with a black star on Fig. 2. The maximum modelled ppm value in the area around the aircraft encounter is 0.7 ppm (4 times above the WHO-based guideline value) at an altitude of 11-11.5 km, which is a typical aircraft cruising altitude (Fig. 2b and c). As most humans are likely to smell SO<sub>2</sub> at around 3-7 times the WHO-based guideline value, this agrees with reported sulphur smells from the pilot reports. However, it should be noted that olfactory perception is influenced by pressure and decreases at high altitudes (Altundağ et al. 2014), but there is a lack of literature on how the olfactory perception level might change in a pressurised air cabin.

A large portion of the modelled  $SO_2$  cloud includes  $SO_2$  concentrations that exceeded the WHO-based guideline as shown by the  $SO_2$  hazard area (the red area) on Fig. 2d. This illustrates the potential hazard area for aviation from a human health perspective due to volcanic  $SO_2$ . Note that this area shows where the  $SO_2$  concentrations exceed the WHO-based guideline value at any vertical model level, i.e., anywhere over the vertical depth of the modelled atmosphere. The area aligns with the grey line in Fig. 2b outlining the 0.175 ppm level.

In this case, the  $SO_2$  hazard area can be compared to the volcanic ash hazard area because the ash and  $SO_2$ clouds from Kasatochi were co-located and travelled together for a period of time (Corradini et al. 2010). The volcanic ash hazard area is taken from the VAA, which is the official volcanic ash forecast issued from Washington



Fig. 1 NAME simulated transport of the volcanic  $SO_2$  clouds from the **a**) Raikoke 2019 eruption and **b**) the 2008 Kasatochi eruption, 4 days after eruption onsets. Modelled  $SO_2$  total column values in Dobson Unit (DU) are shown

VAAC during the time of the eruption. The VAA was based on satellite imagery, model data and pilot reports. By overlaying the VAA polygon (dashed line in Fig. 2d) onto the SO<sub>2</sub> hazard area (red area in Fig. 2d), we see that in this case the areas cover about the same extent, except the VAA does not cover the southernmost part of the modelled SO<sub>2</sub> hazard area, which interestingly has a lower SO<sub>2</sub> plume altitude (Fig. 2c). The VAA area is larger than the SO<sub>2</sub> hazard area, but this is partly due to the fact that the shape of VAA polygon is limited by a set number of vertices and so complex cloud structures cannot be depicted. The area along the Canadian coast where aircraft encounters were reported is also within the VAA polygon.

The  $SO_2$  and ash hazard areas are compared in a similar way for the Raikoke 2019 eruption. Figure 3 shows the Raikoke  $SO_2$  hazard areas at three different times overlaid with the VAA polygons issued by the Tokyo and Anchorage VAACs. In the early hours after the eruption (Fig. 3a) the agreement between the  $SO_2$  and ash hazard areas based on this approach is excellent and illustrates that the

ash and SO<sub>2</sub> were initially transported together. However, over time the agreement diverges as parts of the ash and SO<sub>2</sub> clouds start to separate and travel in different directions due to particle sedimentation, chemistry, and aerosol-radiation interaction (Bruckert et al. 2022). Figure 3b shows a situation where the volcanic ash hazard area is larger than that for SO<sub>2</sub>, however the limited points that can be used to draw the ash polygon make this a simplified representation and prevent a detailed comparison. Later (Fig. 3c), there is very good agreement between the SO<sub>2</sub> and ash area boundaries at the northern edge of the eruption clouds, but overall, the SO<sub>2</sub> hazard area is larger than that for ash.

We now look at the time evolution of the extent of the  $SO_2$  hazard areas when applying the WHO-based guideline (i.e., the red areas in Figs. 2d and 3). The extent changes with time as illustrated in Fig. 4. For the two largest, explosive eruption case studies considered here (Kasatochi and Raikoke), the hazard areas become very large and cover an area up to the size of Alaska (~1.7 million km<sup>2</sup>) and Greenland (~2.2 million km<sup>2</sup>), respectively,



**Fig. 2** The Kasatochi SO<sub>2</sub> cloud at 21:30 UTC 10 August 2008. **a** Modelled total atmospheric column values in Dobson Unit [DU]. **b** Modelled maximum ppm concentration (maximum over altitude). Outer grey line shows the 0.175 ppm contour level. **c** Modelled altitude [km as]] where maximum ppm value occurs. **d** SO<sub>2</sub> hazard area where modelled SO<sub>2</sub> concentrations exceed the WHO-based guideline value (0.175 ppm) at any altitude in the atmosphere (area aligns with grey contour level in **b**). Dashed black line is the VAA polygon at 21:30 UTC extending from surface to 12.2 km altitude (i.e., Flight Level SFC/FL400). The black star marks the approximate location of an aircraft encounter at around 01:00 UTC on 11 August reporting sulphur smells (approximate location taken from Guffanti et al. 2010a, b). The black triangle shows the location of the Kasatochi volcano

after about 4 days. The differences in the hazard areas for the two eruptions are due to differences in the transport patterns rather than the source emissions (Fig. 1, Table 1). The issue time of the last VAA for these two eruptions is shown as the vertical dashed lines on Fig. 4. For both Kasatochi and Raikoke the last VAA was issued on day 5, which implies that after this time volcanic ash was no longer expected to be a hazard for aircraft. However, the SO<sub>2</sub> concentrations exceed the WHO-based guideline value for 1-2 weeks.

The Puyehue, Merapi and Holuhraun eruptions released significant amounts of  $SO_2$  into the atmosphere, but with a smaller  $SO_2$  flux than Kasatochi and Raikoke. The Puyehue  $SO_2$  cloud circulated the Earth three times (the first circuit took 9-10 days) and was detected by satellites for a about three weeks (Clarisse et al. 2012). However, Fig. 4 illustrates that  $SO_2$  concentrations in the Puyehue  $SO_2$  cloud exceeded the WHO-based guideline value for only 3 days. Hence, the part of the modelled  $SO_2$  cloud that reached Australia and further encircled the Earth did not contain  $SO_2$ 

concentrations above the WHO-based guideline. For the Merapi eruption, the WHO-based guideline was exceeded for 4 days and when the SO<sub>2</sub> cloud reached the north Australian coast (as observed by satellite) the guideline was no longer exceeded. These eruptions illustrate that satellites can detect relatively small quantities of SO<sub>2</sub>; however, it is not possible to use nadir sounding satellite data alone to assess whether SO<sub>2</sub> concentrations exceed the WHO-based guideline. This is because a simple relationship does not exist between a volumetric concentration threshold and a satelliteequivalent threshold in Dobson Unit (DU), as the conversion is highly sensitive to the height and thickness of the SO<sub>2</sub> cloud. We note that limb-sounding satellite instruments like the Microwave Limb Sounder (MLS) can estimate vertical profiles of SO2 mixing ratios in the upper troposphere and lower stratosphere (Pumphrey et al. 2015). However, the estimated profiles are spaced 1.5° apart and thus the data has a limited horizontal resolution compared to data from nadir sounding instruments.



Fig. 3 The Raikoke 2019 SO<sub>2</sub> cloud. SO<sub>2</sub> hazard area where modelled SO<sub>2</sub> concentrations exceed the WHO-based guideline value (0.175 ppm) at any altitude in the atmosphere. The dashed black lines are the VAA polygons at different times extending from surface to 13.1 km, 12.2 km and 11.6 km asl (i.e., Flight Levels SFC/FL430, SFC/FL400 and SFC/FL380), respectively



**Fig. 4** Time series of the SO<sub>2</sub> hazard area (where SO<sub>2</sub> concentrations exceed the WHO-based guideline value of 0.175 ppm) for (**a**) all eight eruptions considered in the assessment, and (**b**) a zoom-in for the smaller eruptions. The vertical dashed lines show the issue time of the last VAA for Kasatochi and Raikoke

The  $SO_2$  concentrations related to the effusive Holuhraun eruption exceeded the WHO-based guideline value for smaller areas over a longer period of time compared to the other events. Note that the last VAAs for some eruptions are not included because either the eruption did not release volcanic ash (Holuhraun) and therefore no VAAs exist, or the eruption continued for longer than that simulated and the last VAA is not possible to determine (e.g., for Puyehue over 800 VAAs were issued in 2011 whereas this analysis only considered the major eruptive event).

For Kliuchevskoi and Etna, which are smaller explosive eruptions, the WHO-based guideline is exceeded for up to 24 hours over a relatively small area (up to 25000 km<sup>2</sup> – the size of Sicily), while the effusive eruption of Fogo showed similar SO<sub>2</sub> hazard areas but covering a longer period of time.

To summarise, across the different types of eruptions considered, we find that the period and area over which the WHO-based  $SO_2$  guideline value is exceeded strongly depends on the eruption characteristics: the amount, duration and height of the  $SO_2$  which is released. Eruptions that release large quantities of  $SO_2$  over a short period of time and at high altitudes cause the most extensive  $SO_2$  hazard areas and duration of exceedance. Effusive eruptions that release  $SO_2$  over a significant period of time can lead to prolonged periods of exceedance, but emissions from this type of activity are generally at lower altitudes where  $SO_2$  is removed from the atmosphere more efficiently than at higher altitudes, so the  $\mathrm{SO}_2$  hazard areas are therefore smaller.

We have also applied higher SO<sub>2</sub> concentration thresholds compared to the WHO-based guideline, i.e., exposure to higher concentrations of SO<sub>2</sub> than currently recommended by WHO. Figure 5 shows the difference in the spatial extent of the cumulative SO<sub>2</sub> hazard area exceeding three different SO<sub>2</sub> concentration thresholds (WHO=0.175 ppm, WHOx2=0.35 ppm, WHOx6=1.05 ppm) for the Raikoke and Puyehue eruptions. Note that the cumulated area shows the total cloud area exceeding a given threshold summed over the entire model simulation period and not on a single day. The SO<sub>2</sub> hazard areas and duration of exceedance do not decrease linearly with increasing SO<sub>2</sub> concentration threshold, i.e., a doubling of the threshold does not yield a 50% reduction in cloud extent and duration. By applying the WHOx2 threshold the maximum SO<sub>2</sub> hazard area is reduced by about 40% for these two eruptions, while the number of hours exceeding the threshold is reduced by 26% (Tables 3 and 4). Applying the WHOx6 threshold reduces



**Fig. 5** Cumulative SO<sub>2</sub> hazard area exceeding three different SO<sub>2</sub> concentration thresholds (WHO=0.175 ppm, WHOx2=0.35 ppm, WHOx6=1.05 ppm) for the Raikoke eruption (**a**, **b**, **c**) and the Puyehue eruption (**d**, **e**, **f**). The cumulated area shows the total cloud area exceeding a given threshold summed over the entire model simulation period (and not on a single day)

Descriptor	Etna	Kliuchevskoi	Holuhraun <sup>a</sup>	Fogo <sup>a</sup>	Merapi	Puyehue	Kasatochi	Raikoke
WHO	5100	25177	157085	47563	694526	474896	2332097	1745551
WHOx2	730	5631	73251	10538	197306	226236	1308225	1125841
WHOx6	0	0	17116	956	16087	32297	518181	501852

Table 3 Maximum SO<sub>2</sub> hazard area [km<sup>2</sup>] based on various concentration thresholds

<sup>a</sup> Effusive eruptions with continuous release over many weeks

Table 4 Number of hours that SO<sub>2</sub> concentrations exceed various SO<sub>2</sub> thresholds

Descriptor	Etna	Kliuchevskoi	Holuhraun <sup>a</sup>	Fogo <sup>a</sup>	Merapi	Puyehue	Kasatochi	Raikoke
WHO	22	22	852	284	120	77	186	324
WHOx2	2	11	852	193	79	53	138	238
WHOx6	0	0	700	49	59	31	92	149

<sup>a</sup> Effusive eruptions with continuous release over many weeks

the maximum  $SO_2$  hazard area by about 70-80%, and the number of hours exceeding the threshold is reduced by around 50%. For Merapi and Puyehue, these reductions are even larger. This illustrates that the extent of the  $SO_2$  hazard areas strongly depend on the applied concentration threshold.

# Discussion

We have shown that following a volcanic release of SO<sub>2</sub>, large areas of the atmosphere can contain SO<sub>2</sub> concentrations which are greater than the WHO-based guideline value and as such could pose a health risk to aircraft passengers and crew.

In order to apply the SO<sub>2</sub> concentration thresholds equally at all altitudes in the atmosphere, we must use thresholds in volumetric units (ppm). Our results show a larger aviation impact than that found by Schmidt et al. (2014). They considered a hypothetical short-duration explosive eruption similar to that of Hekla in 2000 emitting 0.2 Tg (200 kt) of SO<sub>2</sub> over 2 hours and found that a 15 min or longer exposure of aircraft and passengers to concentrations  $\geq$ 500 µgm<sup>-3</sup> had a low probability of about 0.1%. One reason for the differences between the two studies is that we have included different eruptions in our analysis, some of which released larger amounts of SO<sub>2</sub> and also over longer periods of time. Secondly, we have applied the WHO guideline in a volumetric unit (ppm), while Schmidt et al. (2014) used a mass concentration threshold in  $\mu gm^{-3}$ , which will vary with height. For example, for an SO<sub>2</sub> plume with initial ground-level concentration of 500 µgm<sup>-3</sup>, the equivalent mass concentration when the plume is at 5 km altitude is 300  $\mu$ gm<sup>-3</sup>, and at 10 km the equivalent mass concentration is ~170  $\mu$ gm<sup>-3</sup> assuming a standard atmospheric pressure gradient. This is due to the atmospheric pressure decreasing with increasing altitude. Therefore, Schmidt's use of 500  $\mu$ gm<sup>-3</sup> at 10 km is equivalent to a volumetric value of about 0.5 ppm (WHOx3).

Our results have shown that SO<sub>2</sub> concentrations above the WHO-based guideline remain in the atmosphere for longer than the time period over which volcanic ash advisories are issued. The reasons for this are likely due to different observational detection limits, different atmospheric lifetimes, as well as different thresholds considered for volcanic SO<sub>2</sub> and ash. It is often easier to detect SO<sub>2</sub> with current satellite instrumentation compared to volcanic ash as the methods for detecting and retrieving physical properties of volcanic ash from satellite observations differ to the methods used for SO<sub>2</sub> (e.g., Theys et al. 2017; Prata et al. 2022). The atmospheric lifetime of SO<sub>2</sub> varies between hours to weeks and depends on factors such as altitude (see Fig. 14 of Carn et al. 2016).  $SO_2$ can have a longer atmospheric lifetime than volcanic ash due to different atmospheric removal processes such as sedimentation, which acts on volcanic ash only. These factors mean that  $SO_2$  can be present and detectable for longer than volcanic ash. There are many examples where volcanic SO<sub>2</sub> clouds have been detected by satellite for days to weeks longer than the corresponding ash cloud (e.g. Prata et al. 2010; de Leeuw et al. 2021; Clarisse et al. 2012).

This study considers the ambient  $SO_2$  concentrations. i.e., outside an aircraft, which may differ to concentrations inside the cabin due to the air exchange or ventilation system onboard. The air exchange system pressurises, heats, and humidifies the ambient air, which at cruise altitudes is very dry and cold, before it enters the aircraft cabin (Bezold 2021). The rate at which air is exchanged will vary between different aircraft types and aircraft ventilation systems. Most systems mix fresh outside air compressed within the engine (known as "bleed air") with recirculated air that has been passed through filters that remove solid and liquid particles in the air (e.g., Burdon et al. 2023). It is outside the scope of this study to evaluate the difference between ambient and in cabin SO<sub>2</sub> concentrations, however it should be accounted for in any further work on establishing the health impact from volcanic SO<sub>2</sub> at flight altitudes. Furthermore, the WHO guideline is applicable to ground air quality, and not directly to aircraft cabin air quality at altitude. Lastly, the WHO guideline used in this study is for a 10 min exposure duration, however the duration of exposure in cabin could be shorter or longer depending on the specifics of the encounter. We lack evidence to suggest whether alternative guidelines would be more appropriate for different exposure durations. Aircraft air quality studies of "fume events" where oils, hydraulic and other fluids contaminate the aircraft bleed air have been extensively documented in the literature (e.g., Burdon et al. 2023), and the use of exposure limits applied to the aircraft cabin environment have been examined (Watterson and Michaelis 2019). However, these studies have not included  $SO_2$ . We recommend that further studies by the relevant agencies assess the medical issue of SO<sub>2</sub> standards within aircraft.

Our analyses are based on model data which include uncertainties. Comparisons between satellite and model results is key to assess these uncertainties. However, a direct evaluation between modelled concentration data and nadir sounding satellite observations is not possible because SO<sub>2</sub> satellite observations are a total atmospheric column quantity. All of the eruption case-studies included in this assessment have been previously studied in detail including thorough validation with independent data (Heard et al. 2012; Schmidt et al. 2014, 2015; de Leeuw et al. 2021; Kristiansen et al. 2010). The eruption source parameters we have used have been taken from studies which have determined them using the best available observations, often in combination with modelling. However, even with thorough validation, there are uncertainties in the results that should be considered. These include uncertainties in the model inputs (both the emissions and meteorology), the model chemistry and other model parameterisations (e.g., mesoscale motions, de Leeuw et al. 2021). The amount, timing, height and vertical distribution of the SO<sub>2</sub> emissions are not exactly known, even when they are derived from satellite data, because different satellite instruments have their own uncertainties and can give different values for the same eruption (e.g., Kristiansen et al. 2010). This study uses a

large range of eruption sizes which provide output that cover a range expected to be largely inclusive of overall source term error. However, errors and uncertainties in the meteorological data can affect the transport and positioning of the SO<sub>2</sub> cloud in the model simulations (e.g., Dacre and Harvey 2018). For example, incorrect representation of vertical wind shear can lead to the SO<sub>2</sub> cloud being transported in the wrong direction. These factors mean that some discrepancies between the model and observed clouds are expected and that uncertainties in the spatial and temporal location of the modelled SO<sub>2</sub> clouds should be considered. The use of ensemble-based model simulations (i.e., meteorology and/or source term ensembles) has in recent years shown promise as a way of representing uncertainty in the modelling framework (e.g., Leadbetter et al. 2022). Lastly, the model output used in our study is average concentrations over the model's horizontal, vertical and temporal grid size. A higher resolution grid size will resolve finer details in the concentrations, however more model particles are needed to accurately compute the concentrations, adding to the computational cost. A coarse model grid size will result in smoother concentration variations. The chosen grid sizes were relatively high resolution and determined by a trade-off between accuracy and computational cost. The uncertainties due to the chosen grid size are thought to be smaller than source term or meteorological uncertainties.

The aviation impact from volcanic SO<sub>2</sub> clouds will depend not only on the concentration threshold applied but also on the height and layer thickness of the SO<sub>2</sub> cloud which we have not explored in detail in this present study. Different styles of eruptions will affect different altitude regions. For example, effusive eruptions can release substantial amounts of SO<sub>2</sub> at low altitude, which may affect airports and impact aircraft during take-off and early climb phases of the flight. Larger eruptions which release SO<sub>2</sub> to higher altitude will affect aviation at cruise altitude. Lidar observations have often showed that aged volcanic clouds typically evolve into thin layers in the atmosphere (e.g., Prata et al. 2015). Therefore, the SO<sub>2</sub> hazard might be localized to certain vertical layers, and the volume of airspace impacted could be smaller than indicated in our present study which has condensed the findings into a 2-D horizontal extent. A volcanic SO<sub>2</sub> forecasting service for aviation would need to provide full 3-D information about the location of SO<sub>2</sub> in the atmosphere to enable appropriate decision making.

# Conclusions

Volcanic  $SO_2$  model simulations for eight historic eruptions have been used to calculate the area and time over which  $SO_2$  concentrations exceed the World Health Organisation (WHO) based guideline value, suggesting a potential health risk for aircraft passengers and crew. The eruptions considered span a large range of eruption styles and sizes, allowing a broad evaluation of types of events that could impact air traffic. The approach used was to apply the WHO SO<sub>2</sub> concentration guideline in volumetric unit (e.g., ppm) at different atmospheric altitudes to evaluate threshold exceedance. A mass concentration threshold (e.g.,  $\mu$ gm<sup>-3</sup>) was not used as it will vary with altitude.

The main findings of this threshold analysis are that:

- The WHO-based guideline value (0.175 ppm) is exceeded for all the historic eruptions considered.
- The time and area over which the WHO-based guideline is exceeded varies and depends on the eruption characteristics i.e., the amount, duration and height of the SO<sub>2</sub> which is released.
- Eruptions which release large quantities of SO<sub>2</sub> at high altitudes have the biggest impact; the most extensive SO<sub>2</sub> hazard areas and longest duration of exceedance.
- For the largest eruptions considered in our analysis, the SO<sub>2</sub> hazard area defined by the WHO-based guideline often covered a considerably larger area than the volcanic ash hazard area defined by the Volcanic Ash Advisories (VAA) issued for the same eruption. Similarly, the SO<sub>2</sub> hazard areas were present for much longer (1-2 weeks) compared to the VAA issue period (a few days). The difference is likely due to different observational detection limits, different atmospheric lifetimes, as well as different thresholds considered for volcanic SO<sub>2</sub> and ash.
- Effusive eruptions which release SO<sub>2</sub> over a long time (weeks to months) can lead to prolonged periods of exceedance, but emissions from this type of activity are generally at lower altitudes where SO<sub>2</sub> is removed from the atmosphere more efficiently than at higher altitudes, and the SO<sub>2</sub> hazard areas are therefore smaller.
- Satellite detection of the SO<sub>2</sub> cloud using nadir sounding instruments is, alone, not sufficient to determine whether the concentration exceeds the WHO-based guideline. SO<sub>2</sub> concentrations in the Puyehue SO<sub>2</sub> cloud exceeded the WHO-based guideline for 3 days, whilst the SO<sub>2</sub> cloud was detected by satellite for a much longer period of time as it encircled the Earth.
- By applying a higher SO<sub>2</sub> concentration threshold (WHOx6) close to the average human SO<sub>2</sub> odour threshold (between WHOx3 and WHOx7) the maximum hazard area is reduced by about 70-80%, and the number of hours exceeding the threshold is

reduced by  $\sim$ 50% for the two largest eruptions considered and the reductions are even larger for the other eruptions.

Our analysis shows that if the WHO-based guideline was introduced in an aviation service, there would be an increase in the frequency and duration of advisory products for volcanic clouds and the areas covered by these products would be much larger than currently issued for volcanic ash. The potential impact on aviation depends on the position of the  $SO_2$  cloud in relation to air traffic corridors and the required response to the advisory. Further work is needed to assess the medical issue of  $SO_2$  standards within aircraft.

### Abbreviations

DU	Dobson Unit
-L	Flight Level
CAO	International Civil Aviation Organisation
MetUM	Met Office's global Unified Model
NAME	Numerical Atmospheric-dispersion Modelling Environment
SO <sub>2</sub>	Sulphur Dioxide
VAA	Volcanic Ash Advisory
VAAC	Volcanic Ash Advisory Centre
NHO	World Health Organisation

# **Supplementary Information**

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Supplementary Material 1.

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### Authors' contributions

Conceptualization, N.I.K., C.S.W.; Funding acquisition, C.S.W.; Investigation, N.I.K., C.S.W., F.M.B.; Methodology, N.I.K., C.S.W.; Software, N.I.K., C.S.W., F.M.B.; Visualization, N.I.K.; Writing—original draft, N.I.K.; Writing—review & editing, N.I.K., C.S.W., F.M.B. All authors have read and agreed to the published version of the manuscript.

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# Availability of data and materials

The NAME model is available for use under license. Model input data in the form of  $SO_2$  source term information for each eruption considered in this work, is provided in the supplementary section.

### Declarations

### Competing interests

The authors declare no competing interests.

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