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Key Points:
- Air quality standards not violated in far field for 2010 Eyjafjallajökull
- Low risk of plume encounters that violate AQ standard for Hekla type
- Predicting the occurrence of volcanic sulfur may aid aviation risk management

Supporting Information:
- Readme
- Figure S1
- Table S1
- Table S2

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Assessing hazards to aviation from sulfur dioxide emitted by explosive Icelandic eruptions

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Abstract Volcanic eruptions take place in Iceland about once every 3 to 5 years. Ash emissions from these eruptions can cause significant disruption to air traffic over Europe and the North Atlantic as is evident from the 2010 eruption of Eyjafjallajökull. Sulfur dioxide (SO_2) is also emitted by volcanoes, but there are no criteria to define when airspace is considered hazardous or nonhazardous. However, SO_2 is a well-known ground-level pollutant that can have detrimental effects on human health. We have used the United Kingdom Met Office's NAME (Numerical Atmospheric-dispersion Modelling Environment) model to simulate SO_2 mass concentrations that could occur in European and North Atlantic airspace for a range of hypothetical explosive eruptions in Iceland with a probability to occur about once every 3 to 5 years. Model performance was evaluated for the 2010 Eyjafjallajökull summit eruption against SO_2 vertical column density retrievals from the Ozone Monitoring Instrument and in situ measurements from the United Kingdom Facility for Airborne Atmospheric Measurements research aircraft. We show that at no time during the 2010 Eyjafjallajökull eruption did SO_2 mass concentrations at flight altitudes violate European air quality standards. In contrast, during a hypothetical short-duration explosive eruption similar to Hekla in 2000 (emitting 0.2 Tg of SO_2 within 2 h, or an average SO_2 release rate 250 times that of Eyjafjallajökull 2010), simulated SO_2 concentrations are greater than 1063 $\mu g/m^3$ for about 48 h in a small area of European and North Atlantic airspace. By calculating the occurrence of aircraft encounters with the volcanic plume of a short-duration eruption, we show that a 15 min or longer exposure of aircraft and passengers to concentrations $\geq$500 $\mu g/m^3$ has a probability of about 0.1%. Although exposure of humans to such concentrations may lead to irritations to the eyes, nose and, throat and cause increased airway resistance even in healthy individuals, the risk is very low. However, the fact that volcanic ash and sulfur species are not always collocated and that passenger comfort could be compromised might be incentives to provide real-time information on the presence or absence of volcanic SO_2. Such information could aid aviation risk management during and after volcanic eruptions.

1. Introduction

Icelandic volcanism features almost all known eruption styles and types, ranging from purely effusive, ash-poor eruptions to purely explosive, ash-dominated eruptions [e.g., *Thordarson and Larsen*, 2007; *Larsen and Eiríksson*, 2008]. The 2010 explosive eruption of Eyjafjallajökull (63.38°N, 19.36°W, 1660 m above sea level (asl)) began on 14 April 2010 and lasted 39 days resulting in severe disruption to air traffic due to the repeated presence of ash plumes in European and North Atlantic airspace [e.g., *Schumann et al.*, 2011; *Gudmundsson et al.*, 2012; *Stevenson et al.*, 2012]. The eruption's impacts quickly reached global scale, affecting several industry sectors [e.g., *Budd et al.*, 2010; *Mazzocchi et al.*, 2010; *Donovan and Oppenheimer*, 2011; *Harris et al.*, 2012] despite its low magnitude of explosivity (volcanic explosivity index of 3) and a total tephra volume of 0.18 km^3 (dense rock equivalent) [*Gudmundsson et al.*, 2012]. Controlled European airspace was restricted for commercial air traffic during 15–23 April 2010 followed by intermittent restrictions of parts of European airspace in the weeks thereafter. These restrictions resulted from the combination of frequent and persistent northwesterly air flow at the altitude at which significant amounts of fine-grained volcanic ash particles were injected (3–10 km) and the aviation safety protocols in place at the time (i.e., "zero tolerance") [e.g., *Gudmundsson et al.*, 2012].

Airborne volcanic ash is a well-recognized hazard to jet engine aircraft [e.g., *Bernard and Rose*, 1990; *Casadevall*, 1994a; *Casadevall et al.*, 1996; *Miller and Casadevall*, 2000; *Prata and Tupper*, 2009] as evident from more than 90 documented encounters since 1953, nine of which resulted in temporary engine shutdown [*Casadevall*, 1994a, 1994b; *Casadevall et al.*, 1996; *Guffanti et al.*, 2010]. Since Eyjafjallajökull 2010, there has been a research focus on atmospheric dispersion modeling of volcanic ash (including the prediction of quantitative ash concentrations) and ground- and space-borne remote and in situ measurements of volcanic ash [e.g., *Clarisse et al.*, 2010; *Flentje et al.*, 2010; *Zehner*, 2010; *Arason et al.*, 2011; *Gislason et al.*, 2011; *Schäfer et al.*, 2011; *Schumann et al.*, 2011; *Stohl et al.*, 2011; *Thomas and Prata*, 2011; *Carboni et al.*, 2012; *Christopher et al.*, 2012; *Gudmundsson et al.*, 2012; *Johnson et al.*, 2012; *Prata and Prata*, 2012; *Rauthe-Schöch et al.*, 2012; *Rix et al.*, 2012; *Stevenson et al.*, 2012; *Turnbull et al.*, 2012; *Webster et al.*, 2012; *Weinzierl et al.*, 2012; *Winker et al.*, 2012; *Boichu et al.*, 2013; *Dacre et al.*, 2013; *Pappalardo et al.*, 2013].

Volcanic plumes not only consist of ash but also contain species such as sulfur dioxide (SO_2) and hydrogen sulfide (H_2S). Generally, SO_2 is considered hazardous to aircraft frames and engines only following its oxidation to sulfuric acid, which upon hydration forms sulfuric acid aerosol particles [*Bernard and Rose*, 1990], typically composed of 75 wt % H_2SO_4 and 25 wt % H_2O [e.g., *Hamill et al.*, 1977]. Sulfuric acid aerosol particles can cause damage to the airframe and/or engine components as a result of sulfidation, leading to corrosion of nickel alloys in gas turbines if alkali metal salts are copresent (e.g., sea-salt, mineral dust) [*Eliaz et al.*, 2002]. Sulfuric acid particles have the potential to cause corrosion and erosion of compressor blades and other engine components [*Swadźba et al.*, 1993]. This may necessitate more frequent maintenance cycles for aircraft and therefore result in increased total cost of ownership.

A few aircraft encounters with volcanic plumes containing SO_2 and/or sulfuric acid aerosol have been documented [*Bernard and Rose*, 1990; *Casadevall et al.*, 1996]. These incidences include reports of distress to pilots and passengers during encounters with the 2011 Grímsvötn volcanic SO_2 plume [*European Space Agency*, 2011] and the Sarychev 2009 plume [*Guffanti et al.*, 2010], but there are no reported cases of damage to aircraft engines or avionics due to encounters with volcanic SO_2 plumes. In contrast to volcanic ash hazards to aviation, there are no criteria to define when airspace is considered hazardous or nonhazardous for volcanic SO_2 plumes. There is, however, International Civil Aviation Organization guidance for flight crew upon noticing the smell of sulfur in an aircraft cabin, and it is recommended that the pilot quickly contact air traffic control and the airline operations center for any information about relevant volcanic activity [*International Civil Aviation Organization*, 2004; *International Volcanic Ash Task Force (IVATF)*, 2012].

At present, none of the Volcanic Ash Advisory Centres (VAACs) worldwide is required to forecast the dispersion or concentration of volcanic SO_2 or sulfuric acid aerosol, although VAACs have started to utilize satellite retrievals of volcanic SO_2 to inform their volcanic ash forecasts [*Tupper et al.*, 2004; *Brenot et al.*, 2014]. In future, the requirements to monitor and forecast SO_2 might change for several reasons. *Carn et al.* [2009] discussed that monitoring and tracking volcanic SO_2 plumes can be a useful proxy for discernable volcanic ash although ash and SO_2 are not necessarily collocated [e.g., *Schumann et al.*, 2011; *Thomas and Prata*, 2011; *Sears et al.*, 2013]. *Carn et al.* [2009] also discussed that cumulative effects of multiple exposure of aircraft to long-lived and aged volcanic ash and aerosol plumes may result in increased cost of ownership. Furthermore, sulfurous odors can cause distress of cabin passengers and aircrew [*Guffanti et al.*, 2010].

Sulfur dioxide has been measured in volcanic plumes at concentrations much higher than those considered toxic to humans [e.g., *Baxter*, 2000; *Rose et al.*, 2003; *Hansell and Oppenheimer*, 2004; *Hunton et al.*, 2005; *Voigt et al.*, 2014]. Many of these measurements were taken at ground level and in close proximity to the volcanic vent, but in situ measurements reported by *Rose et al.* [2003] suggest SO_2 concentrations of 500–1000 ppbv during an aircraft encounter with a 35 h old volcanic plume from the Icelandic Hekla eruption in February 2000 at about 1300 km distance from the source. For context, humans may perceive SO_2 at about 300 ppbv, and the World Health Organization (WHO) sets a 10 min mean ambient air quality standard of 500 $\mu g/m^3$ (about 190 ppbv at ground level) to protect public health [*World Health Organization (WHO)*, 2014]. International Volcanic Ash Task Force, which was established following the 2010 Eyjafjallajökull eruption recommended that for volcanic SO_2, further "work on identifying and quantifying any associated health risks to aircraft occupants […] with a view to enhancing the existing guidance […]" was needed [*IVATF*, 2012]. This area of research has now been taken up by the International Airways Volcano Watch Operations Group [e.g., *International Airways Volcano Watch Operations Group*, 2014].

Table 1. Eruption Scenarios and Their Eruption Source Parameters Used for the NAME Model Simulations[a]

Eruption Scenario	SO_2 Mass Released [Tg SO_2]	Maximum SO_2 Release Height (km asl)	Average and Peak Rate of SO_2 Release [Tg SO_2/h]	Period Considered for Analysis	Historic Eruption Example	References
Long-duration (39 days) Benmoreite to trachyte magma composition.	0.39	Varies between 2.9 and 9.5	Average = 0.0004 Peak = 0.003	14 Apr to 30 May 2010	14 Apr to 22 May 2010 Eyjafjallajökull	*Arason et al.* [2011]; *Thordarson et al.* [2011]; *Gudmundsson et al.* [2012]; *Petersen et al.* [2012]: See also Table S1.
Intermediate-duration (3 days) Assuming basaltic magma composition.	0.102	Varies between 1.2 and 10.0	Average = 0.0018 Peak = 0.0073	14 to 30 Apr 2010	1 to 5 Nov 2004 Grímsvötn	*Sigmundsson et al.* [2004], and references therein; *Vogfjörd et al.* [2005]; *Jude-Eton et al.* [2012]; *Oddsson et al.* [2012]
Short-duration (2 h) Assuming intermediate magma composition.	0.2	12.0	Average = 0.1 Peak = 0.1	14–19 Apr 2010	26 Feb 2000 Hekla	*IES* [2000]; *Rose et al.*, [2003]; *Lacasse et al.* [2004]; *Höskuldsson et al.* [2007]; *Moune et al.* [2007]

[a]The SO_2 mass fluxes for the long-duration and the intermediate-duration eruption scenarios are given in Tables S1 and S2 in the supporting information.

We use the United Kingdom Met Office's NAME (Numerical Atmospheric-dispersion Modelling Environment) model [*Jones et al.*, 2007] to simulate SO_2 mass concentrations for a range of hypothetical explosive Icelandic eruptions. Our aim is to inform the aviation industry and regulators on the likely SO_2 mass concentrations that aircraft could encounter over Europe and the North Atlantic. The eruption case studies span eruption magnitudes, durations, and SO_2 fluxes that are representative of past explosive eruptions in Iceland with a probability to occur about once every 3 to 5 years. The eruption source parameters and the probability are derived from the record of volcanism in Iceland over the last 1150 years [*Thordarson and Larsen*, 2007] and further supported by the Holocene eruption record in Iceland [*Larsen and Eiríksson*, 2008; *Óladóttir et al.*, 2008; *Thordarson and Höskuldsson*, 2008; *Óladóttir et al.*, 2011].

2. Methods

2.1. Model Setup and Definition of the Eruption Case Studies

The NAME model is a Lagrangian dispersion model [*Jones et al.*, 2007], which is used for a range of applications such as air quality modeling and forecasting and emergency response work predicting the dispersion of hazardous nuclear, volcanic, chemical, or biological material [e.g., *Webster et al.*, 2006; *Derwent et al.*, 2007; *Redington et al.*, 2009; *Leadbetter and Hort*, 2011; *Leadbetter et al.*, 2012]. NAME is also used by the London VAAC to forecast volcanic ash dispersion operationally [*Webster et al.*, 2012; *Witham et al.*, 2012]. Previously, *Heard et al.* [2012] used NAME to simulate SO_2 and sulfuric acid aerosol mass concentrations from the eruptions of Kasatochi in 2008, Sarychev in 2009, and Eyjafjallajökull in 2010. These authors found good agreement between satellite-retrieved and model-simulated SO_2 column densities and sulfuric acid aerosol optical depths, which gives confidence in the model's ability to capture not only atmospheric dispersion of volcanic gases and aerosol particles but also their atmospheric column densities.

To provide atmospheric SO_2 mass concentration estimates for a range of explosive volcanic eruptions in Iceland, we simulate (1) Eyjafjallajökull 2010, which is representative of a long-duration explosive eruption with sustained activity for 39 days releasing 0.39 Tg of SO_2 in total, (2) a hypothetical short-duration (2 h) explosive volcanic eruption releasing 0.2 Tg of SO_2 by mimicking the eruption of Hekla in 2000 in Iceland, and (3) a hypothetical intermediate-duration eruption releasing a total of 0.1 Tg of SO_2 over 3 days by mimicking the eruption of Grímsvötn in 2004 (Table 1). The SO_2 release rates for the long-duration and intermediate-duration eruptions vary over the course of the eruption with average hourly release rates of 0.0004 Tg and 0.0018 Tg (Table 1; see also Tables S1 and S2 in the supporting information). For the short-duration eruption, the hourly SO_2 release rate is 0.1 Tg.

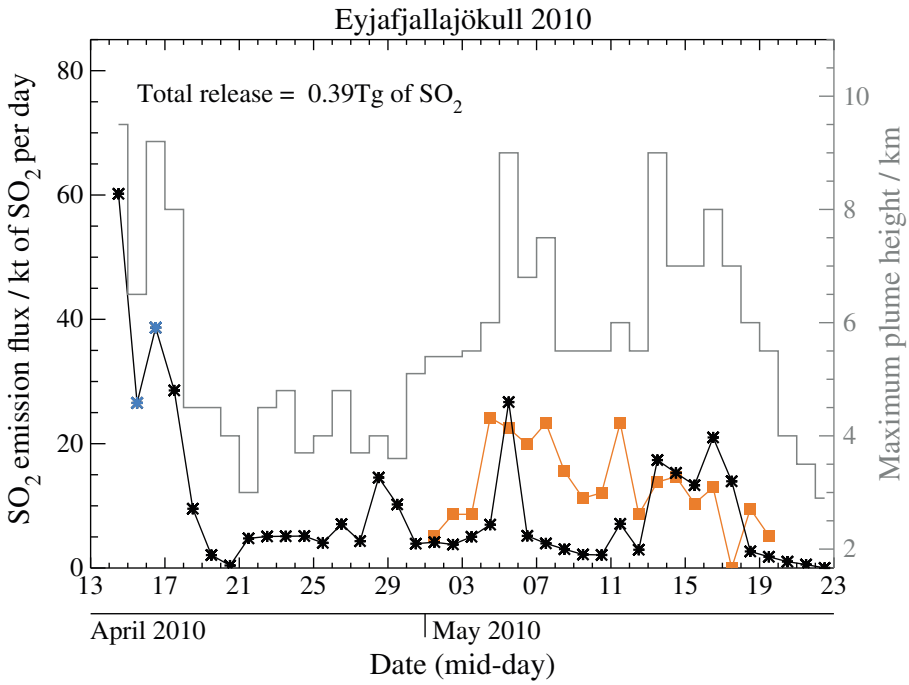


Figure 1. Daily sulfur dioxide (SO_2) mass fluxes (black stars and axis labels on the left, in kilotons per day) for the 2010 Eyjafjallajökull eruption derived using the petrological method [*Thordarson et al.*, 2011] and daily maximum plume height (solid grey line and axis labels on the right, in kilometer above sea level) used in the model to simulate the long-duration eruption scenario. Blue stars indicate that in the model, the SO_2 mass flux was reduced to 0.1% of the petrological flux estimate in order to account for in-plume scavenging of sulfur species as observed during the early phases of Eyjafjallajökull 2010. Orange squares are SO_2 mass fluxes derived using GOME-2 retrievals in combination with inverse modeling [*Flemming and Inness*, 2013].

The eruption scenarios we define are generic but representative of small to moderate magnitude explosive eruptions in Iceland [*Thordarson and Larsen*, 2007; *Óladóttir et al.*, 2008; *Thordarson and Höskuldsson*, 2008; *Óladóttir et al.*, 2011]. The scenarios are not tied to a specific volcano or a specific volcanic system in Iceland because the exact location, magnitude, style, and composition of the next eruption in Iceland are not foreseeable. For example, the next Hekla eruption may be of a smaller or larger magnitude than simulated here and could produce magma of basaltic, intermediate or silicic composition. Since the total mass of SO_2 released is related to magma composition and eruption size (i.e., magma volume), the SO_2 mass flux to the atmosphere may be significantly higher or lower than simulated in our study.

To simulate a long-duration eruption, we use daily varying SO_2 mass fluxes derived from the petrological analysis of the sulfur content in melt inclusions and quenched eruption products for the 2010 summit eruption at Eyjafjallajökull [*Thordarson et al.*, 2011]. The SO_2 mass fluxes are used together with daily plume height data obtained from radar measurements in Keflavik, Iceland [*Arason et al.*, 2011; *Gudmundsson et al.*, 2012], as inputs for NAME (Figure 1). The daily maximum plume height represents the top height of the SO_2 release, and in the model, SO_2 is released uniformly between the Eyjafjallajökull volcano summit (1666 m asl) and the plume top. To account for in-plume scavenging of SO_2 as observed during the early phase of Eyjafjallajökull 2010, we reduced the SO_2 mass flux used in the model to 0.1% of the petrological flux estimate (blue stars, Figure 1). Figure 1 additionally shows SO_2 mass fluxes derived by *Flemming and Inness* [2013] using inverse modeling and GOME-2 (Global Ozone Monitoring Experiment-2) satellite retrievals for the period 1 to 19 May 2010. For the period 6 to 12 May 2010 the SO_2 flux derived using GOME-2 is up to a factor of 5.9 greater than the SO_2 mass flux we derived using the petrological method (Figure 1). These differences demonstrate that satellite-derived fluxes and petrological fluxes can differ substantially, which contributes directly to uncertainties in the model to data comparison.

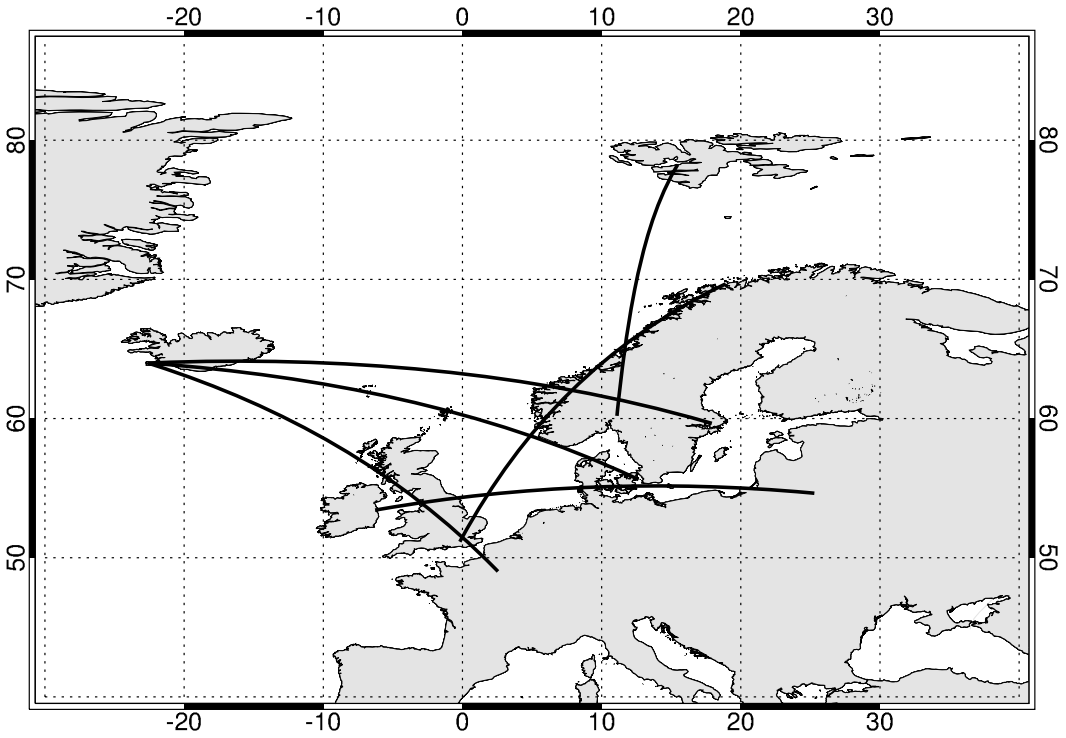


Figure 2. Map showing the five representative flight paths (black lines) that are used to calculate the probability of 15 min or longer periods that an aircraft could be exposed to volcanic sulfur dioxide. These paths are defined so that they pass through the volcanic plume of each eruption scenario.

For all simulations reanalyzed meteorological fields (ERA-interim) from the European Centre for Medium-Range Weather Forecasts [*Dee et al.*, 2011] are used for the period 14 April 2010 to 30 May 2010. We used the same meteorological data for all eruption scenarios, and each eruption commenced midday on 14 April 2010 because under these meteorological conditions SO_2 will be dispersed into European and North Atlantic airspace [e.g., *Petersen et al.*, 2012]. All simulations are run accounting for oxidation of SO_2 as well as dry and wet removal of SO_2 and its oxidation products [*Webster and Thomson*, 2011, 2014]. Details of the NAME chemistry scheme are described in *Heard et al.* [2012] and *Redington et al.* [2009]. Briefly, we simulate the gas-phase reaction of SO_2 with the hydroxyl radical (OH), which is the dominant sink of volcanic SO_2 in the free troposphere. Aqueous-phase oxidation of SO_2 by hydrogen peroxide and ozone occurs in grid boxes where both liquid water and cloud fraction are nonzero [*Redington et al.*, 2009; *Heard et al.*, 2012]. Mass concentrations of SO_2 are output as 15 min means on a regular longitude-latitude grid of 0.5° by 0.5°.

We apply a similar procedure as the London VAAC applies for volcanic ash to our analysis of SO_2 mass concentrations at flight altitudes in European and North Atlantic airspace by outputting mass concentrations for 2500 ft deep layers [see also *Webster et al.*, 2012; *Witham et al.*, 2012]. Flight levels (FLs) are barometric pressures expressed as a nominal altitude in 100 s of feet. For the analysis, we combined the 2500 ft deep output to three FL ranges by finding the maximum mass concentration in the following FL ranges: FL000-FL200, FL200-FL350, and FL350-FL550. The cruising altitude of commercial aircraft on an intercontinental flight lies between 20,000 ft and 35,000 ft, equivalent to FL200-FL350. The cruising altitude on transatlantic flights typically lies between 35,000 ft and 45,000 ft (FL350-FL450). FL000-FL200 is relevant for low-altitude cruising and for takeoff and landing.

2.2. Ozone Monitoring Instrument Satellite Retrievals and In Situ Measurements of SO_2

The retrieval of SO_2 vertical columns densities (VCDs, in Dobson units, DU) from Ozone Monitoring Instrument (OMI) is achieved by applying Differential Optical Absorption Spectroscopy (DOAS) [*Platt and Stutz*, 2008] to the ultraviolet measured spectra in the 312–326 nm wavelength range. An empirical offset

correction is then applied to account for possible biases and is estimated over clean regions. The obtained quantity is converted into an SO_2 VCD using an air mass factor that accounts for changes in measurement sensitivity due to observation geometry, ozone column, clouds, and surface reflectivity. The SO_2 VCD is estimated with the assumption of a plume height at 7 km above sea level and is provided along with the column operator, which accounts for the altitude-dependent sensitivity of the OMI measurements. Figure S1 shows that the SO_2 VCDs retrieved using the DOAS algorithm are in close agreement with those retrieved using the operational OMI algorithm described in *Krotkov et al.* [2006].

To compare simulated SO_2 VCDs with those retrieved by OMI, vertical model output resolutions of 100 m in the first 1 km and 500 m between 1 km and 16 km have been used. In order to directly compare model output and satellite retrievals, the OMI column operators (accounting for instrument sensitivity in the vertical) have to be applied to the model profiles by taking each OMI pixel and finding the corresponding model profile which is coincident in both time (within the model 15 min mean time window) and space (within a model grid box). The model profile is then interpolated to the OMI vertical retrieval grid, resulting in a simulated VCD that can be directly compared to OMI.

2.3. Calculation of Aircraft Encounters With Volcanic SO_2 Along Flight Paths

To calculate the probability of 15 min or longer periods that an aircraft could be exposed to SO_2, we define five flight paths that are representative of actual flight paths along great circles in North Atlantic and European airspace (Figure 2). These flight paths are defined so that they pass through the volcanic plume of each eruption scenario. For simplicity, we assume that the aircraft flies with a constant ground speed of 700 km/h along these flight paths. The probability of aircraft transiting through or aircraft occupants being exposed to volcanic SO_2 concentrations $\geq X_i$ for 15 min or longer is given as the mean probability (in percent) for the five flight paths with the sample space defined by the analysis period considered for each eruption scenario (Table 1).

3. Results

3.1. Model Evaluation for the 2010 Eyjafjallajökull Eruption

To evaluate the ability of NAME to simulate SO_2 VCDs and the dispersion of SO_2, we compare simulated SO_2 VCDs for the long-duration Eyjafjallajökull 2010 eruption with satellite retrievals from OMI. Additionally, we compare simulated mass mixing ratios of SO_2 to in situ measurements from the United Kingdom's Facility for Airborne Atmospheric Measurements (FAAM) research aircraft [*Johnson et al.*, 2012; *Turnbull et al.*, 2012].

Figure 3 compares the spatial distribution of simulated and measured SO_2 VCDs for 4 days during Eyjafjallajökull 2010 applying the column operators that assume a plume height of 7 km, which best reflects the conditions during the eruption. Qualitatively, the presence or absence of volcanic SO_2 can be captured using NAME as evident from comparing retrieved and simulated SO_2 VCDs. However, NAME underpredicts maximum SO_2 VCDs compared to OMI with a normalized mean bias of −13.6% (Figures 3 and 4a). The relative frequency distributions of all simulated and retrieved SO_2 VCDs greater than 0.5 DU (Figures 4b and 4c) and the relative frequency the bias (NAME minus OMI) further demonstrate that NAME most frequently underpredicts SO_2 VCDs by up to 3 DU compared to OMI (Figure 4d).

The daily maximum SO_2 VCDs retrieved by OMI during April and May 2010 (Figure 4a) are in good agreement with those reported by *Rix et al.* [2012] using GOME-2. For context, a SO_2 VCD of 10 DU converts to mixing ratios on the order of 300 ppbv when assuming a plume thickness of 1 km. The SO_2 VCDs for Eyjafjallajökull 2010 can be put in context with those experienced in airspace during previous volcanic eruptions worldwide. During the 12–17 June 2009 eruption of Sarychev Peak (Kuril Islands, Russia, emitting 1.2 Tg of SO_2), large areas of the Northern Hemisphere experienced SO_2 VCDs greater than 0.5 DU with maximum daily-mean VCDs of about 120 DU retrieved by IASA (Infrared Atmospheric Sounding Interferometer) [*Heard et al.*, 2012]. During the eruption of Kasatochi (Aleutian Islands emitting 1.5 Tg of SO_2) on 7–8 August 2008, satellite-retrieved maximum SO_2 VCDs ranged between 100 and 700 DU depending on the satellite instrument and the assumed plume height [*Clarisse et al.*, 2012, and references therein].

Between 14 May and 18 May 2010, the UK FAAM aircraft was airborne for a total of about 34.5 h. About 0.2% of this time, mixing ratios greater than 75 ppbv have been encountered, which is in close agreement with the fraction of time (0.1%) the German Falcon aircraft encountered mixing ratios greater than 75 ppbv (U. Schumann,

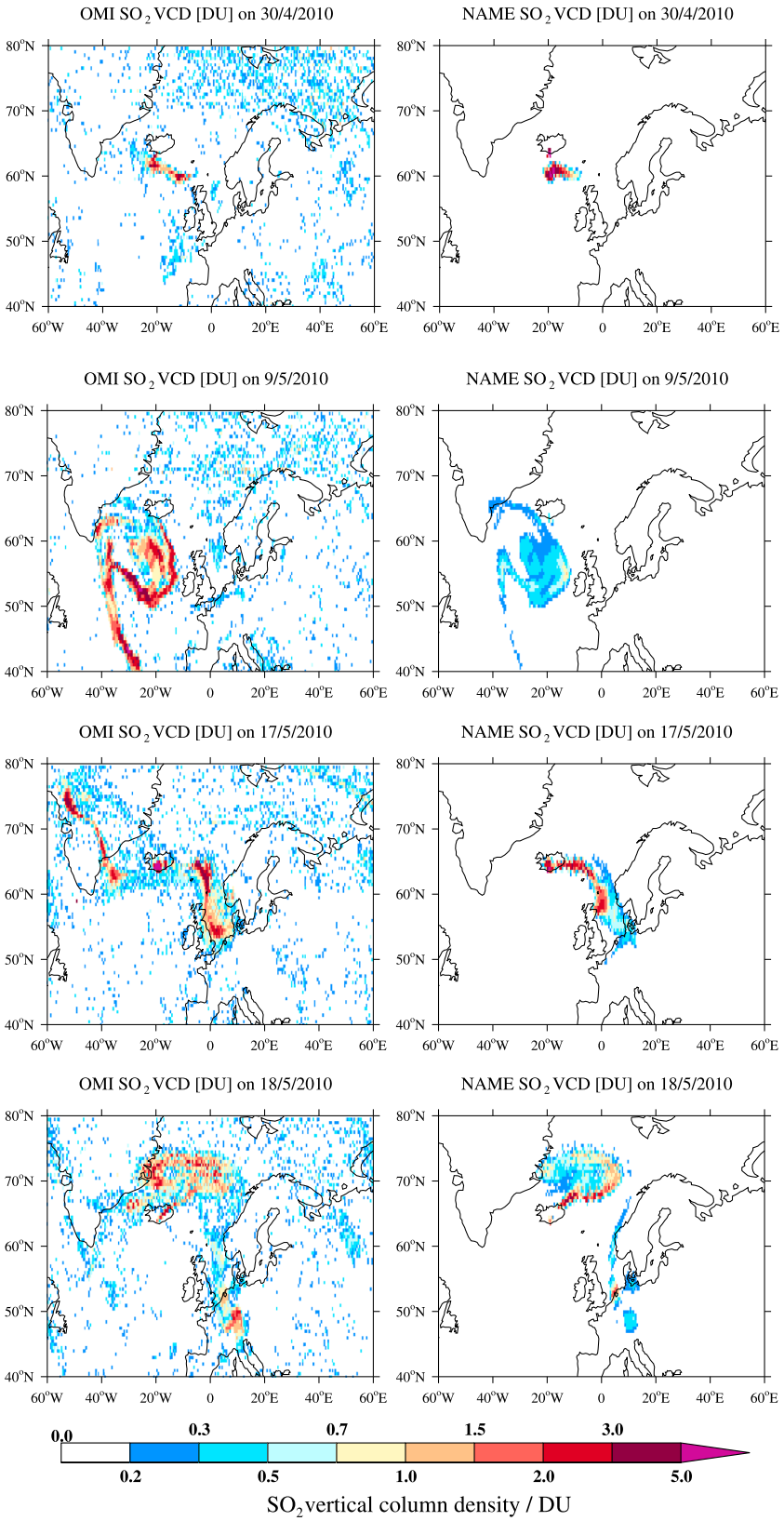


Figure 3. Comparison of the spatial distribution of SO_2 vertical column densities (VCDs in Dobson units, DU) during 4 days of the 2010 Eyjafjallajökull eruption using retrievals from the Ozone Monitoring Instrument (OMI; left) and simulated VCDs from the NAME model (right).

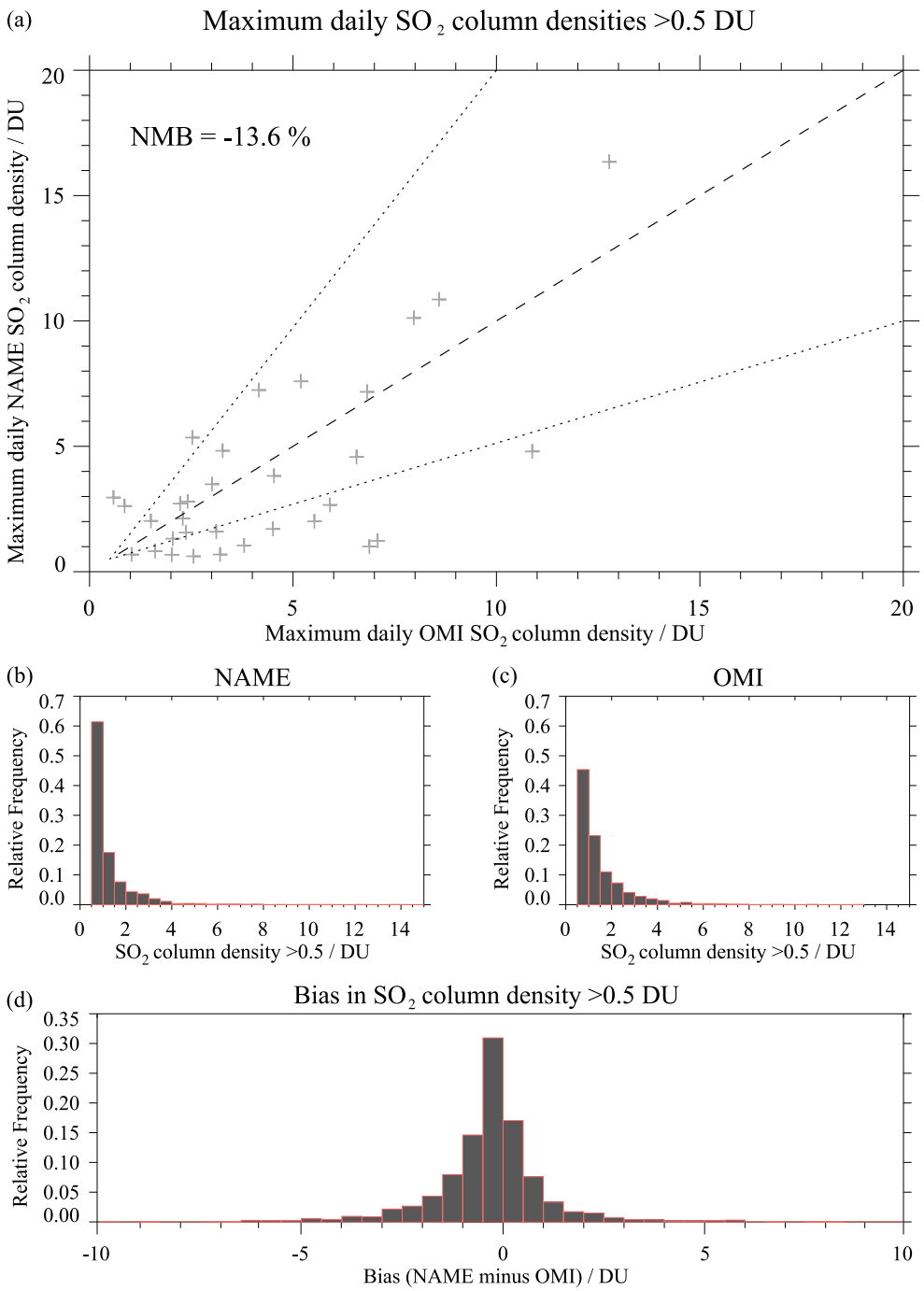


Figure 4. Comparison of (a) maximum SO_2 vertical column densities (VCDs in Dobson units, DU) greater than 0.5 DU retrieved by the Ozone Monitoring Instrument (OMI) with maximum NAME VCDs for the period 14 April to 30 May 2010 of the Eyjafjallajökull eruption. Also shown is the normalized mean bias (NMB in percent). For the comparison, the maximum was located anywhere in a given scene (i.e., maximum VCDs shown may not be spatially collocated in NAME and OMI scenes). The relative frequency distribution of all gridded SO_2 VCDs greater than 0.5 are shown (b) for NAME and (c) for OMI. and (d) The relative frequency distribution of the bias (NAME minus OMI) in SO_2 VCDs.

personal communication, 2014). For Eyjafjallajökull, the simulated maximum SO_2 mass mixing ratios are up to a factor of about 4 lower than those measured by the FAAM research aircraft on 18 May 2010, although there is good agreement of the temporal occurrence of peak concentrations (Figure 5a). Spatially,

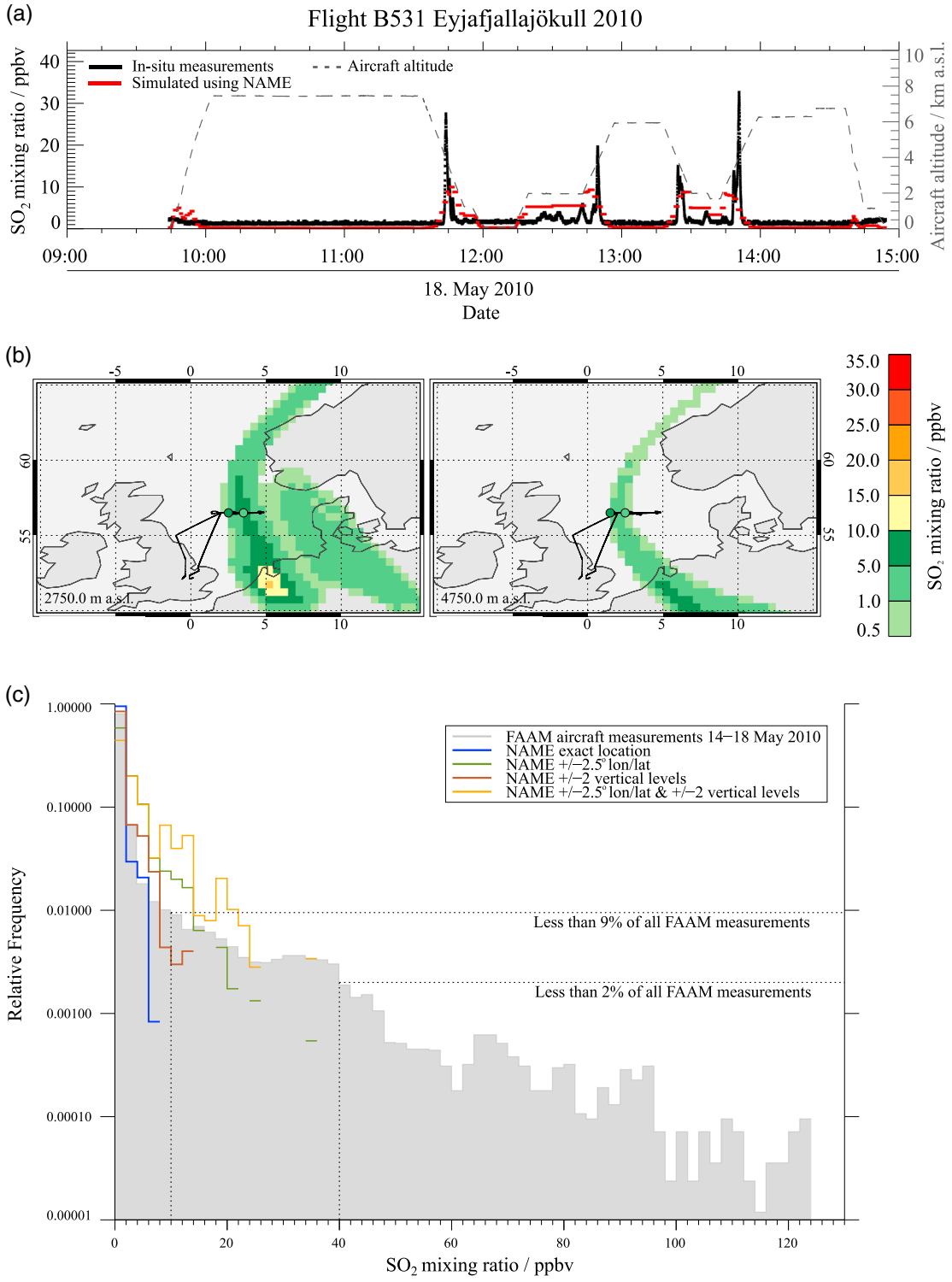
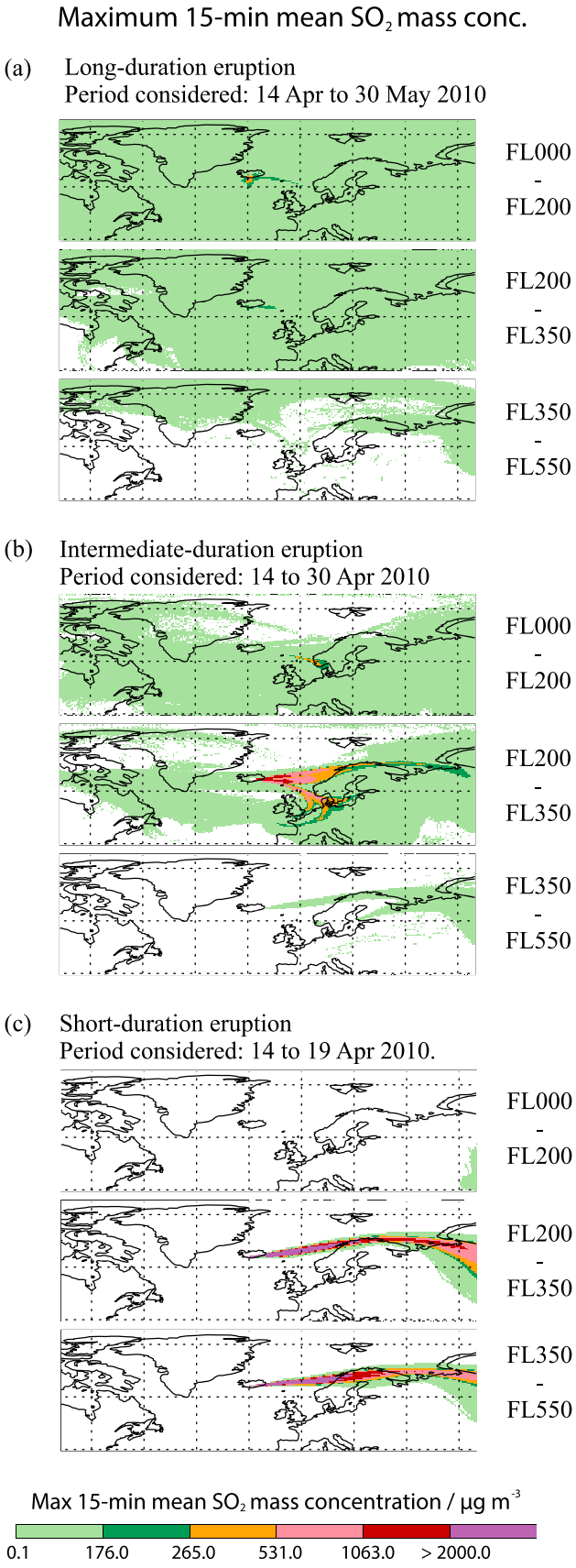


Figure 5. Comparison of SO_2 mixing ratios (in parts per billion by volume, ppbv) measured by the UK FAAM research aircraft during May 2010 to those simulated in NAME. (a) In situ measurements from FAMM flight B531 (black) on 18 May 2010 with simulated (red) SO_2 mixing ratios, which are sampled along the flight track (solid black line in Figure 5b). In Figure 5a the aircraft altitude is shown by the dashed grey line. (b) A comparison of the spatial distribution of simulated SO_2 mixing ratios with in situ measurements (colored circles along flight track) on 18 May 2010. (c) The relative frequency of all aircraft measurements made in the period 14–18 May 2010 (grey) with that of simulated SO_2 mixing ratios, which have been sampled along the FAAM flight paths for the same period (colored lines). The colored lines show simulated mixing ratios obtained using different levels of model sampling tolerance with regard to the flight track (blue = exact location in terms of longitude, latitude, and height above sea level; green = maximum SO_2 mixing ratio found within ±2.5° longitude/latitude; brown = maximum SO_2 mixing ratio found within ±2 vertical output levels (±1 km); orange = maximum SO_2 mixing ratio found within ±2.5° longitude/latitude and ±2 vertical output levels).



simulated SO_2 mixing ratios are in good agreement with FAAM measurements throughout most vertical levels although there are mismatches, which are particularly pronounced for mixing ratios greater than 10 ppbv (Figure 5b). Comparing FAAM aircraft in situ measurements for the period 14–18 May 2010 to the simulated SO_2 mixing ratios, it is apparent that maximum SO_2 mixing ratios are not captured by the model, although in situ measurements of SO_2 mixing ratios greater than 40 ppbv represent less than 2% of all measurements (Figure 5c). Mixing ratios below 40 ppbv are captured by the model if up to ±2.5° tolerance in terms of longitude and latitude and/or ±2 vertical levels (±1 km) tolerance is used with respect to the location of the in situ measurements (Figure 5c, orange and green lines).

3.2. Sulfur Dioxide Mass Concentrations at Flight Altitude and Threshold Exceedances

Based on the UK Daily Air Quality Index defined by the Department for Environment, Food and Rural Affairs (DEFRA), 15 min mean ground-level SO_2 concentrations greater than 265 $\mu g/m^3$ would result in a public health warning being issued [*Connolly et al.*, 2013]. The WHO health protection guideline for SO_2 mass concentrations is set at 500 $\mu g/m^3$ for an averaging period of

Figure 6. Spatial distribution of maximum 15 min mean SO_2 mass concentrations (in $\mu g/m^3$) simulated at flight altitudes at any time over a certain period of time considered for (a) the long-duration eruption, (b) the intermediate-duration eruption, and (c) the short-duration eruption scenario (see section 2 and Table 1 for details on the eruption period considered). Humans can perceive SO_2 at mixing ratios >300 ppbv [*IVHHN*, 2014], which is about 240 $\mu g/m^3$ at 11 km (~36,000 ft) altitude using the U.S. 1976 standard atmospheric profile. The label used for the SO_2 mass concentrations uses a similar color coding as used by the UK Department for Environment, Food, and Rural Affairs (DEFRA) for SO_2 to distinguish warning levels to protect public health [*Connolly et al.*, 2013].

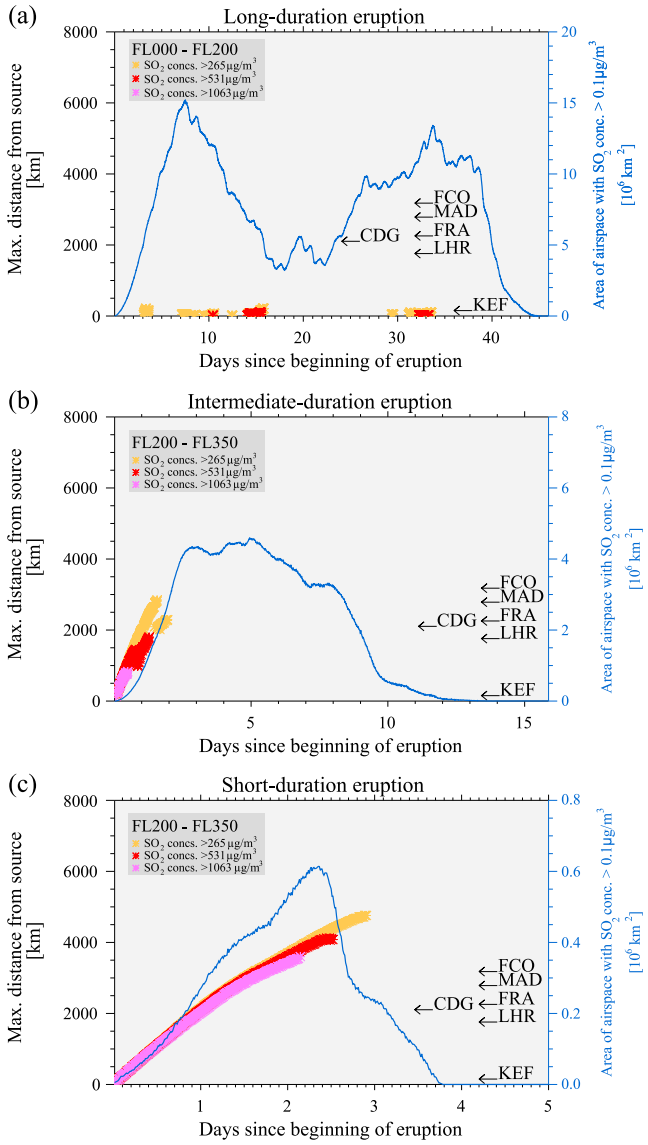


Figure 7. Maximum distance from the volcanic vent a certain threshold SO_2 mass concentration (in $\mu g/m^3$) is exceeded at flight altitudes over the course of each eruption scenario (colored stars; axis label on the left; flight altitude indicated using FL notation) for (a) the long-duration eruption at FL000-FL200, (b) the intermediate-duration eruption at FL200-FL350, and (c) the short-duration eruption at FL200-FL350. The SO_2 mass concentration thresholds are defined based on the UK Department for Environment, Food, and Rural Affairs (DEFRA) legislation and warning levels for SO_2 to protect public health [*Connolly et al.*, 2013]. Humans can perceive SO_2 at mixing ratios >300 ppbv [*IVHHN*, 2014], which is about 240 $\mu g/m^3$ at 11 km (~36,000 ft) altitude using the U.S. 1976 standard atmospheric profile. Also shown are the distances of European airports with respect to the volcanic vent (FCO = Fiumicino Airport, Italy; MAD = Adolfo Suárez Madrid-Barajas Airport, Spain; FRA = Frankfurt Airport, Germany; CDG = Paris Charles de Gaulle Airport, France; LHR = London Heathrow Airport, United Kingdom; KEF = Keflavík International Airport, Iceland). The solid blue line shows the area of airspace where volcanic SO_2 concentrations are greater than 0.1 $\mu g/m^3$ (axis label on the right; area is given in 10^6 km^2 out of about 100×10^6 km^2 of the total area considered in the model domain) for each eruption scenario.

10 min [*WHO*, 2014]. On a 15 min mean basis, SO_2 concentrations greater than 531 $\mu g/m^3$ and 1063 $\mu g/m^3$ are considered "high" and "very high" pollution by DEFRA.

Figure 6 shows the spatial distribution of maximum 15 min mean SO_2 mass concentrations simulated at any time for each eruption case study in the three flight altitude ranges (see section 2 and Table 1 for details). For the long-duration Eyjafjallajökull 2010 eruption, we find that maximum SO_2 mass concentrations do not

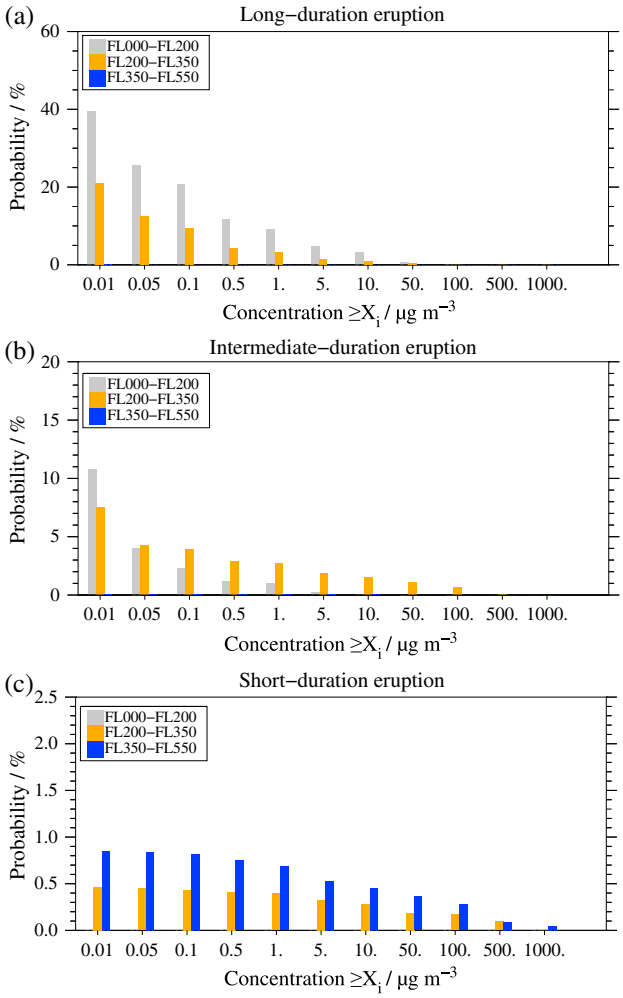


Figure 8. Mean probability (in percent) of aircraft transiting through volcanic SO_2 concentrations $\geq X_i$ (in $\mu g/m^3$) at flight altitudes (grey = FL000-FL200; orange = FL200-FL350; blue = FL350-FL550) for 15 min or longer sampled along any of the five flight paths (shown in Figure 2) for (a) the long-duration eruption, (b) the intermediate-duration eruption, and (c) the short-duration eruption scenario. The sample space used to calculate the probabilities is defined by the analysis period considered for each eruption scenario (Table 1).

exceed 265 $\mu g/m^3$ in any of the three FL ranges (Figure 6a), except in a small region around Iceland. In contrast, during a short-duration explosive eruption emitting 0.2 Tg of SO_2 over a 2 h period (Table 1), maximum SO_2 mass concentrations exceed 1063 $\mu g/m^3$ in FL200-FL350 and FL350-FL550 (Figure 6c). The 1063 $\mu g/m^3$ threshold is exceeded as far downwind of the volcanic vent as ~3100 km for a period of just over 2 days (Figure 7c). Overall, during a short-duration explosive eruption, volcanic SO_2 (with concentrations > 0.1 $\mu g/m^3$) is present in a relatively small area of airspace of about 0.6 million km^2 out of about 100×10^6 km^2 of total area considered in the model domain. For this eruption scenario, SO_2 concentrations exceed 265 $\mu g/m^3$ for at least 3 days in FL200-FL350 and FL350-FL550, which based on the UK DEFRA air quality legislation would result in a public health warning being issued if exceeded at ground level [*Connolly et al.*, 2013]. For the intermediate-duration eruption (emitting a total of ~0.1 Tg of SO_2 over 3 days), SO_2 mass concentrations are greater than 531 $\mu g/m^3$ in FL200-FL350 up to about 1900 km from the volcanic vent, and the 265 $\mu g/m^3$ threshold is exceeded as far downwind as 2900 km during the first 2 days of such an eruption (Figures 6b and 7c). Figure 7 also shows the distances of several international airports in Europe highlighting that during a short- or intermediate-duration eruption, SO_2 concentrations greater than 1063 $\mu g/m^3$ could occur as far away as London Heathrow (LHR, United Kingdom) and Frankfurt (FRA, Germany) about 24 h after the onset of an eruption.

The SO_2 concentrations simulated for each eruption scenario can be compared with in situ measurements made during the Hekla eruption in the year 2000. *Rose et al.* [2003] measured SO_2 volume mixing ratios up to 1000 ppbv at 11.3 km altitude (using the U.S. 1976 standard atmospheric profile, 1000 ppbv is equivalent to about 770 $\mu g/m^3$ at about 11 km), which is of the same order as the maximum SO_2 mass concentrations of about 1050 $\mu g/m^3$ that we find in the 36 h old plume in FL350-FL550 for the short-duration eruption.

3.3. Probability of 15 min or Longer Exposure to SO_2 Along Flight Paths

For the long-duration Eyjafjallajökull 2010 eruption, we find that long continuous encounters of high SO_2 concentrations are unlikely. We find that the maximum concentration that an aircraft would encounter for 15 min or longer is 120 $\mu g/m^3$ (see section 2 and Figures 2 and 8). Figure 8 shows that there is a ~40% probability of exposure for 15 min or longer to concentrations $\geq$0.01 $\mu g/m^3$ and of about 5% of exposure to concentrations $\geq$5 $\mu g/m^3$ along the flight paths in FL000-FL200 (Figure 8a). For the intermediate-duration eruption, there is a 0.7% probability of exposure for 15 min or longer to concentrations $\geq$100 $\mu g/m^3$ in FL200-FL350, and the probability of encountering concentrations $\geq$500 $\mu g/m^3$ is about 0.05% in FL200-FL350 (Figure 8b). For the short-duration eruption scenario, we find that SO_2 mass concentrations $\geq$500 $\mu g/m^3$ could be encountered for 15 min or longer in FL200-FL350 or FL350-FL550, although the probability of such an encounter is just under 0.1% (Figure 8c). The probability of encountering concentrations $\geq$ 1000 $\mu g/m^3$ for 15 min or longer in FL350-FL550 is about 0.01%.

A study on SO_2 infiltration into aircraft cabins commissioned by the Department for Transport in the United Kingdom suggests that SO_2 concentrations inside a narrow body aircraft (e.g., Boeing 757-300 or Airbus 320) will reach about 80% of the ambient atmospheric concentrations within less than 3 min upon encounter [*Chitty and Shipp*, 2013]. This is valid for ambient volume mixing ratios that exceed 200 ppbv for SO_2 (equivalent to ~182 $\mu g/m^3$ at 10 km altitude), and the clearance time required for in-cabin concentrations to return to "zero" is about 12 min [*Chitty and Shipp*, 2013]. Although the probability of encountering SO_2 concentrations greater than 500 $\mu g/m^3$ for 15 min or longer is lower than 0.1% for the short-duration eruption scenario, such an encounter would violate public health protection legislation set for ground-level pollution. This may be relevant even more if the cabin air clearance time is taken into account.

4. Discussion

A handful of other studies and aviation safety protocols have suggested that more research on volcanic SO_2 may be useful for assessing aviation hazards [*Carn et al.*, 2009; *Bonadonna et al.*, 2012, 2014; *IVATF*, 2012; *Rix et al.*, 2012]. However, data on likely SO_2 concentrations at flight altitude for Icelandic eruptions are scarce and there are no hazard assessments available.

For the Eyjafjallajökull eruption in 2010, we show that the spatial distribution of simulated SO_2 VCDs compare qualitatively well with those retrieved by OMI (Figures 3 and 4). The comparison gives confidence in the suitability of the London VAAC NAME dispersion model as a tool to monitor and to predict the presence or absence of volcanic SO_2, which may be a useful quantity to aid aviation risk management. However, simulating SO_2 concentrations at exact point locations or integrating concentrations along flight paths (i.e., dosage) is more challenging and subject to large uncertainties, which is a similar issue in quantitative volcanic ash concentration forecasting [e.g., *Rauthe-Schöch et al.*, 2012; *Webster et al.*, 2012].

In order to quantitatively predict SO_2 mass concentrations, several sources of uncertainty have to be taken into account such as the accuracy of the eruption source terms (plume height and SO_2 mass flux), meteorological fields, chemical conversion rates, gas and aerosol removal rates, as well as the limitations of the vertical, horizontal, and temporal model and output resolution (Figure 5) and the representation of in-plume processes near the source in the model. For our model simulation of the 2010 Eyjafjallajökull eruption, we have accounted for in-plume scavenging of SO_2 during the early phases of the eruption in a very simplistic way by reducing the SO_2 mass flux to the atmosphere to 0.1%. Uncertainties in meteorological fields used in the model appear to be minor given the reasonable representation of the spatial occurrence of the volcanic plume compared to in situ measurements and OMI satellite retrievals. However, in general, the farther away from the volcanic vent, the greater the errors on plume concentrations and plume location tend to become in models (see *Boichu et al.* [2013] for a detailed discussion). In terms of SO_2 mass fluxes, we

estimate an uncertainty on the petrological SO_2 mass fluxes of about ±20%. When converting satellite SO_2 VCDs to mass fluxes, uncertainty arises from the assumptions made about the chemical conversion rates of SO_2 to form aerosol particles [e.g., *Carn et al.*, 2013; *Flemming and Inness*, 2013].

There are no aviation safety regulations for volcanic SO_2; therefore, we have used air quality and health protection guidelines defined for ground-level pollution in order to assess the hazards due to volcanic SO_2 at flight altitude. Based on this approach, we conclude that at no time during the long-duration Eyjafjallajökull eruption in 2010 did 15 min mean SO_2 mass concentrations in the far field (≥1000 km away from the volcanic source) significantly exceed air quality standards set to protect public health even under the assumption that NAME underpredicts maximum SO_2 concentrations. However, in a small area (less than 200 km^2) over and around Iceland, SO_2 concentrations exceeded 265 $\mu g/m^3$ on several days in our model (Figures 6a and 7a), which based on the UK DEFRA air quality legislation would result in a public health warning being issued [*Connolly et al.*, 2013]. In contrast, during a short-duration explosive eruption with an eruptive phase lasting 2 h (Table 1, with a SO_2 release rate 250 times the average release rate of the long-duration Eyjafjallajökull 2010 eruption scenario), SO_2 mass concentrations greater than 1063 $\mu g/m^3$ occur for about 48 h up to about 3500 km away from the volcanic vent (Figure 7c). Concentrations greater than 1063 $\mu g/m^3$ are considered "very high pollution" by DEFRA if exceeded at ground level [*Connolly et al.*, 2013]. For the short-duration eruption scenario, SO_2 concentrations farther than ~1500 km from the volcanic vent reach up to ~3000 $\mu g/m^3$ in both FL200-FL350 and FL350-FL550 (Figure 6c).

On the basis that in-cabin concentrations reach 80% of the ambient atmospheric concentrations within three minutes and about 12 min of cabin air clearance time [*Chitty and Shipp*, 2013], our results suggest that passengers in an aircraft encountering volcanic SO_2 from a short-duration eruption as simulated here could be exposed to concentrations greater than 500 $\mu g/m^3$ for 15 min or longer. Although exposure of humans to SO_2 concentrations greater than ≥500 $\mu g/m^3$ for 10 min or longer may lead to irritations to the eyes, nose and throat, and cause increased airway resistance even in healthy individuals [*Baxter*, 2000; *World Health Organization*, 2000; *EPA*, 2008; *International Volcanic Health Hazard Network (IVHHN)*, 2014; *WHO*, 2014], the probability of such a plume encounter in North Atlantic airspace is as low as 0.1% (Figure 8c). Most likely, such an encounter would result in passenger comfort being compromised (rather than in detrimental health effects) since individuals may perceive sulfur odors at these concentrations.

5. Summary and Implications

At present, there is no defined level of susceptibility of aircraft, avionics or aircraft passengers to volcanic SO_2, which limits the assessment of the hazards and risks. If aviation regulator and airline operators consider information on the presence or absence of SO_2 as useful for aiding aviation risk mitigation, then models like NAME could be used to provide these information. Airborne volcanic ash clearly poses a greater threat to aircraft operations [e.g., *Guffanti et al.*, 2010] than volcanic SO_2 for those eruption scenarios we have assessed. However, the fact that volcanic ash and sulfur species are not always collocated [e.g., *Schumann et al.*, 2011; *Thomas and Prata*, 2011; *Sears et al.*, 2013] or that passenger comfort could be compromised might be further incentives to forecast SO_2 concentrations operationally in addition to volcanic ash. Probabilistic assessment of the SO_2 hazards from explosive volcanism in Iceland similar to those carried out for volcanic ash [*Biass et al.*, 2014] would be possible if regulators and the aviation industry were to define levels of susceptibility.

Quantitative forecasting of volcanic SO_2 concentrations or their integrated quantities along flight paths (i.e., dosage) would be much more challenging than merely providing information on the presence or absence of volcanic SO_2. This is mainly because accurate eruption source terms at high temporal resolution (SO_2 mass flux, height of the emissions, and eruption characteristics such as near-source scavenging of sulfur species) are required as model inputs. Providing near-real-time data on the SO_2 flux is challenging, and we have shown that there can be significant differences between satellite-derived source terms and those derived using petrology (see section 2 and Figure 1). These differences are mainly a result of specific eruption characteristics like scavenging of sulfur species in the volcanic plume [e.g., *Sigmarsson et al.*, 2013], but also the fact that satellites retrieve column density instead of fluxes although techniques are developed to convert these into mass fluxes [e.g., *Carn et al.*, 2013; *Flemming and Inness*, 2013; *Theys et al.*,

2013]. In an operational context, assimilating ground-based, aircraft, and satellite measurements of volcanic SO_2 would help to reduce uncertainties in model forecasts of volcanic SO_2 [*Brenot et al.*, 2013; *Flemming and Inness*, 2013; *Boichu et al.*, 2014], but these techniques have not yet been fully developed for operational use.

We have assessed the SO_2 hazard for eruption case studies of explosive volcanism in Iceland that are representative of small to moderate magnitude explosive eruptions. These case studies have been defined based on the record of eruptions in Iceland in the last 1150 years [e.g., *Thordarson and Larsen*, 2007]. From this record, it also is known that there have been at least four long-lasting (months to years) flood lava eruptions that produced magma volumes greater than 1 km^3. For the 1783–1784 A.D. Laki eruption, for example, the average hourly SO_2 release rate during the first 3 months [*Thordarson et al.*, 1996; *Thordarson and Self*, 2003] was up to 3 orders of magnitude greater than that during Eyjafjallajökull in 2010. On the basis of our results for explosive eruptions, and previous modeling studies of a Laki-type eruption [*Schmidt et al.*, 2010, 2011; *Schmidt*, 2015], sulfur species from infrequent but very large-scale flood lava eruptions like Laki may present a major challenge for aviation risk management depending on volcanic plume heights, SO_2 release rates, and the duration of volcanic activity.

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