# UK Hazards from a Large Icelandic Effusive Eruption

# **Effusive Eruption Modelling Project Final Report**

Project workers:



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This report was prepared under contract for the Civil Contingencies Secretariat (Cabinet Office) H55 Project Board. The contents of this report have undergone review by the H55 Project Board and H55 Scientific Expert Advisory Group, convened by the Civil Contingencies Secretariat.

The report is published by the Met Office on behalf of the Civil Contingencies Secretariat.

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This publication is available for download at: www.metoffice.gov.uk

Witham, C., Aspinall, W., Braban, C., Hall, J., Loughlin, S., Schmidt, A., Vieno, M., Bealey, B., Hort, M., Ilyinskaya, E., Kentisbeer, J., Roberts, E., Rowe, E., 2015, UK Hazards from a Large Icelandic Effusive Eruption, Effusive Eruption Modelling Project Final Report.

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# **Executive Summary**

In response to the recent introduction of large, long-lasting gas-rich volcanic eruptions to the UK National Risk Register (risk H55) a modelling project has been conducted to improve our understanding of potential hazards to the UK from such an eruption on Iceland. A precautionary "reasonable worst case" eruption scenario based on the 1783-1784 CE Laki eruption has been determined using the results of an expert elicitation of scientists. This scenario has been simulated 80 times using two different atmospheric chemistry and transport models (NAME and EMEP4UK) over 10 years of meteorology (2003-2012).

The results provide information on the range of concentrations of sulphur dioxide  $(SO_2)$ , sulphate aerosol  $(SO_4)$  and some halogen species that might be experienced in the UK during such an eruption and the likelihood of key thresholds being exceeded and the duration of their exceedance. Data for the surface and for a range of key flight altitudes have been produced. These are evaluated against the threshold bandings of the UK's Air Quality Index (AQI). The impact on UK ecosystems has also been considered. The data are intended to be used by UK Government Departments for further research into the impacts on the aviation, health, environmental and agricultural sectors.

The results show that the prevailing meteorological conditions are the key influence on which parts of the North Atlantic and European region are affected at any time. The results demonstrate that the UK is unlikely to be affected by week after week of significantly elevated concentrations; rather there will a number of short (hours to days) pollution episodes where concentrations at the surface would be elevated above Moderate and High air quality index levels. This pattern reflects the generally changeable nature of the weather in the UK. At the surface, consecutive exceedance durations are longer for SO<sub>4</sub> than SO<sub>2</sub>, and can be particularly lengthy (1-2 weeks) in the Low air quality index levels, which may be of relevance to health impact assessments.

The indications of potential peak concentrations and their corresponding AQI exceedance probabilities within this report serve to inform national, high-level generic risk planning. For more specific response planning, a much larger modelling study with multiple eruption scenarios and a greater number of meteorological realisations would be needed.

#### **Eruption Source Term**

An eruption source term for  $SO_2$  emissions from a 'Laki-type' eruption has been defined for use in evaluating the extent to which  $SO_2$  and  $SO_4$  might be hazardous at the ground and at flight levels over the UK during a future eruption.

- Relevant eruption source parameters (emission height, duration, etc) were estimated using the findings of an expert elicitation.
- Sequences of daily eruption source terms extending over a period of 6 weeks were then produced using stochastic modelling.
  - These include information on plume height and SO<sub>2</sub> mass erupted for a five-layer height profile for each time period, which allows variations in emission mass with height and time to be accounted for.
- One sequence, representing a "precautionary worst case" for the UK, has been selected for use in the modelling. In this scenario, approximately 24% of the  $SO_2$  is emitted into the stratosphere and 76% is emitted into the troposphere over the course of the eruption.

• Start dates for eighty, non-overlapping, 6-week eruption periods in 2003-2012 have been defined. Model simulations were then conducted for each of these periods using both NAME and EMEP4UK.

#### Near Surface Results

Results from the model simulations conducted with NAME and EMEP4UK for  $SO_2$  and  $SO_4$  at the near-surface show that:

- Surface level SO<sub>4</sub> will be present mostly as partly or fully neutralised ammonium sulphate particles rather than sulphuric acid.
- Median and 95<sup>th</sup> percentile concentrations of SO<sub>2</sub> and SO<sub>4</sub> are within the *Low* AQI levels in both models for both hourly and daily (24-hour) average concentrations.
- A gas and aerosol plume can arrive at the surface in the UK (Northern Ireland) as quickly as 29 hours after the start of the eruption, but generally takes several days.
- The prevailing meteorological conditions are very important and influence which parts of the UK are affected at any time, if the UK is affected at all.
- The maximum percentage of simulations in which SO<sub>2</sub> concentrations reached *Moderate* air quality levels at least once anywhere at the surface over the UK and Republic of Ireland is ~16% for hourly averages and ~3% for daily averages. For SO<sub>4</sub> the maximum percentages are ~95% for hourly averages and ~44% for daily averages.
- The maximum percentage of simulations in which SO<sub>2</sub> concentrations entered into *High* air quality levels anywhere at the surface is ~5% for hourly averages and 0% for daily averages. For SO<sub>4</sub> the maximum percentages are ~84% for hourly averages and ~32% for daily averages.
- Once exceeded, the duration of exceedance tends to be relatively short (i.e. hours to days). The maximum consecutive duration at *High* air quality levels for hourly averages found in any of the NAME simulations was <15 hours for SO<sub>2</sub> and <114 hours for SO<sub>4</sub>.
- The NAME results suggest that *Very High* concentrations of SO<sub>2</sub> may occur at the surface, although these periods are rare in the model simulations (occurring in only 2 simulations out of 80 in the hourly data only) and short-lived (a few hours only). EMEP4UK does not simulate values above High for SO<sub>2</sub>.
- For SO<sub>4</sub>, both NAME and EMEP4UK show that *Very High/Hazardous* concentrations of SO<sub>4</sub> may occur. The maximum consecutive duration is 2-3 days for the UK, which is much longer than that for SO<sub>2</sub>.
- On average over a 6-week eruption period concentrations are elevated above background at the surface for only a short number of days/hours: less than 5 hours for SO<sub>2</sub> and less than 6 days for SO<sub>4</sub> (both based on hourly averages).
- On average the south of the UK experiences longer pollution events than Scotland.

#### Flight Altitude Results

Results from the model simulations conducted with NAME and EMEP4UK for  $SO_2$  and  $SO_4$  at aviation flight levels show that for hourly data:

- Median and 95<sup>th</sup> percentile concentrations from both models are within the *Low* air quality index levels for both species at all flight levels over the UK.
- To the east of Iceland (extending as far east as the Faroe Islands at upper flight levels) 95<sup>th</sup> percentiles of SO<sub>2</sub> are in the *Moderate* index level in both models. This distribution is characteristic of the prevailing wind conditions near Iceland.

- In all of the model simulations concentrations of SO<sub>2</sub> reached the *Moderate* AQI level at some point at all cruising and on-route flight levels over the UK.
- At higher altitudes (24,000 ft and above) the chance of exceeding  $SO_2$  thresholds increases. This is because most of the gas is emitted from the volcano at these heights. Greater concentrations of  $SO_4$  are more likely at lower altitudes.
- The maximum consecutive duration of exceedance of *High* air quality levels for SO<sub>2</sub> is much greater at flight altitudes than the surface. The maximum duration at cruise and on-route altitudes (24,000-37,000 ft) over the UK in any of the simulations was 95 hours for SO<sub>2</sub> and 31 hours for SO<sub>4</sub>.
- The consecutive duration of elevated levels of  $SO_4$  increases at lower flight altitudes. Maximum consecutive durations at *High* air quality levels at 7,000 ft are 93 hours for  $SO_2$  and 94 hours for  $SO_4$ .
- Arrival times over the UK can be as little as 6-7 hours from the start of eruption at upper flight levels. This is much quicker than at the surface due to transport within the jet stream. Over these short time-frames the plumes contain mainly SO<sub>2</sub>, as conversion to SO<sub>4</sub> is still occurring.
- At flight levels the plumes are very transient with their location driven by the meteorology.
- Elevated concentrations do not necessarily occur in all flight levels at the same times reflecting the complex four-dimensional evolution of the volcanic plumes in transit to and over the UK.

#### Impact on UK habitats

The average annual impact of the scenario eruption on ecosystems in the UK has been derived based on EMEP4UK simulations conducted for 2005. This shows that:

- Annual sulphur deposition in the UK increases by roughly 50%, to levels similar to the peak of industrial sulphur pollution in the early 1970s.
- This results in the area of sensitive ecosystems exceeding their critical load (and thus implying potential damage to the ecosystem) doubling, from 22% to 51% of the total area of sensitive ecosystems in the UK exceeding these values.
- The magnitude of the critical load exceedance more than doubles, although it remains relatively low compared to existing geographical variation in exceedances.
- Dwarf shrub heath is most at risk of adverse impacts, followed by montane and bog habitats. Scotland experiences the greatest proportional increase in critical load exceedance amongst all UK countries largely because the majority of these types of habitats exist there.
- Annual average SO<sub>2</sub> gaseous concentrations across the UK increase, although not significantly. Therefore the area exceeding the critical levels of 10  $\mu$ g/m<sup>3</sup> and 20  $\mu$ g/m<sup>3</sup> remains the same as in non-eruption years.
- The average pH across the UK drops (becomes more acidic) only slightly but returns to near normal within five years. However, even a small drop in pH can result in damage when pH is already low, particularly to freshwater natural systems and aquaculture. Such impacts are likely be localised.
- The effects of concentrated, short-term peaks of sulphur deposition could not be fully evaluated using the yearly model applied in the study. However, the modelling suggests that periods of high sulphur concentrations in the nearsurface atmosphere would be brief and short-term and severe acidification episodes are therefore unlikely.

#### Hazards from halogen gases and hydrogen sulphide

An assessment of the hazards to the UK from the halogen acids, hydrogen chloride (HCI) and hydrogen fluoride (HF), and hydrogen sulphide (H<sub>2</sub>S) has been conducted.

- A limited literature survey has been carried out on measurements of the three species in volcanic plumes.
- Conservative initial emission ratios of these species with respect to SO<sub>2</sub> were assessed to be: HCI:SO<sub>2</sub> = 1, HF:SO<sub>2</sub> = 0.3, H<sub>2</sub>S:SO<sub>2</sub> = 0.04.
- These ratios were used in EMEP4UK modelling to assess possible hazards to the UK.
- HCl or HF emissions of the same magnitude as SO<sub>2</sub> result in average concentrations at the UK surface of mostly <1  $\mu$ g/m<sup>3</sup> with a 95<sup>th</sup> percentile of <5  $\mu$ g/m<sup>3</sup>. Maximum hourly concentrations are of the order 200-300  $\mu$ g/m<sup>3</sup>. The hourly human health limits of 500  $\mu$ g/m<sup>3</sup> and 750  $\mu$ g/m<sup>3</sup> respectively are not exceeded in these simulations.
- H<sub>2</sub>S also does not exceed the human health 24-hour limit (150 μg/m<sup>3</sup>). Daily average values are generally <2 μg/m<sup>3</sup> with maximum modelled values of ~30 μg/m<sup>3</sup>.
- The results are only indicative as to the likely impact of such an event. There are large uncertainties for all three species considered and therefore without further studies at volcanoes and fundamental parameters being measured in the laboratory, this type of analysis is only an approximation.

#### Uncertainties

The extent to which the results within this study capture the range of variability in the hazard is subject to a large number of factors. These include the choice of eruption source terms, the physics and chemistry of the models and the meteorology. Differences between the results from the two models are evident, as would be expected in any model comparison, and indicate some of the variability. The large uncertainty that these factors introduce must be considered when interpreting the results and for impact assessments. A detailed uncertainty assessment was beyond the scope of this work, but a limited model sensitivity study and a stochastic statistical analysis have been conducted to investigate some of the uncertainty in the source term.

Seven NAME model simulations (using a simplified but plausible scenario) have been conducted to quantify the impact of changing the mass of  $SO_2$  emitted, the height range of the emissions and the duration of emission. These changes introduce considerable uncertainty to near-surface concentrations of  $SO_2$  and  $SO_4$ .

- The height of emission in relation to the tropopause has a first order effect on the near-surface concentrations of both species. Emissions into the stratosphere only reach the surface in small amounts.
- Concentrations of SO<sub>2</sub> have a linear relation to changes in emitted mass over the range tested, so if the emission mass is doubled, and all else held constant, then the downwind concentrations also double. Concentrations of SO<sub>4</sub> do not vary in this linear way.
- Maximum concentrations at the surface vary by a factor of ~9 for SO<sub>2</sub> and ~30 for SO<sub>4</sub> across these sensitivity simulations.
- SO<sub>4</sub> concentrations remain elevated at the surface much longer than SO<sub>2</sub> in all cases.
- The area affected by a given AQI threshold value can vary by a factor of 3, with higher mass emissions and longer release times both contributing to larger areas being affected at the surface and at flight altitudes.

A simplified stochastic variational technique has been applied to some key factors (eruption magnitude and wind speed) that could influence peak volcanic gas concentrations over the UK during a future eruption. Findings from one case study period suggest that:

- The 90% credible interval for plausible peak near-surface SO<sub>2</sub> concentration, somewhere over the UK, is a range from 1300 to 3600  $\mu$ g/m<sup>3</sup>. This implies potential for localized effects associated with *Very high* to *Very hazardous* AQI levels.
- There is a small, but non-zero probability that quite large areas of the UK might, in one episode, experience SO<sub>2</sub> concentrations corresponding to AQI *High* Band 9 or higher; however the overall countrywide peak concentration level is unlikely to exceed *Low*.
- Even a relatively small fissure eruption on Iceland, emitting gas into low altitudes for long enough, could affect the UK.

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# 1 Introduction

Following the impacts of the Eyjafjallajokull eruption in 2010, two types of volcanic eruption have been added to the UK Government's National Risk Register for Civil Emergencies. One of these, a large gas-rich "effusive" volcanic eruption in Iceland, was identified as a high impact natural hazard, and initial assessments based on limited scientific information placed this as one of the three highest priority natural hazards faced by the UK. This eruption scenario is typified by the Laki eruption in 1783-1784. Since 2010, the Civil Contingency Secretariat (CCS) of the UK's Cabinet Office, responsible for Civil Protection in the UK, has been working on quantifying the risk from such an eruption and better understanding its potential impacts. This involves cross-cutting work across UK Government departments and the wider scientific community in order to identify the capabilities needed to respond to an effusive eruption, to exercise the response and develop increased resilience where possible.

The Effusive Eruption Modelling Project (EEMP) was initiated in 2013 to produce a modelling study that will improve our understanding of potential impacts to the UK from such an effusive gas-rich eruption on Iceland.

The results presented in this report provide information on the range of concentrations of sulphur dioxide  $(SO_2)$ , sulphate aerosol  $(SO_4)$  and some halogen species that might be experienced in the UK during such an eruption and the likelihood of key thresholds being exceeded and over what durations. The work represents a detailed initial study but has not explored the full range of such an eruption and has not considered any volcanic ash emissions from an effusive eruption. The outputs from this study are intended to be used by the respective Government Departments for further area-specific impact assessments for sectors including: health, aviation and environment.

## 1.1 Previous Work

The 1783-84 fissure eruption of Laki is considered the "type" eruption for a longlasting effusive eruption on Iceland. The Laki eruption dynamics and emission masses have been characterised in several papers (Thordarson and Self, 1993; 2003) and these have formed the basis for previous assessments of the potential gas and aerosol cloud from such an eruption (e.g. Chenet et al. (2005), Highwood and Stevenson (2003), Schmidt et al. (2011), Schmidt et al. (2012), Stevenson et al. (2003), Oman et al. (2006)). Building on this, in 2012 the Civil Contingencies Secretariat requested that the source characteristics of a future Laki-type eruption scenario be considered in more detail to facilitate an assessment of the potential impacts on the UK. An expert elicitation was proposed as the best way to establish preliminary values for eruption characteristics and their uncertainties, and in April 2012 the British Geological Survey brought together a multidisciplinary group of experts to conduct this elicitation. Scientists were asked to consider a fissure eruption of similar duration to the 1783-1784 Laki eruption (8 months) and to give probability distributions for parameters such as plume height, magma production rates and SO<sub>2</sub> mass release. By capturing and formally combining the group's informed judgements, the expert elicitation allows exploration of the range of possibilities based on current knowledge. The outcomes of the elicitation are presented in Loughlin et al (2013) along with a description of the eruption and its impacts based on published research. These data have been used to define a suitable eruption scenario for the work in this report (see Section 3).

Initial modelling work was conducted in the Spring/Summer of 2013 by the Met Office and University of Leeds for the Civil Contingencies Secretariat H55 Project Board. This considered 11 short eruption periods from April to August 2010 with a limited 6hour eruption of SO<sub>2</sub>, where the emissions from each eruption were followed for 14 days. The full methodology and results were provided to the H55 Project Board as a PowerPoint presentation at the Project Board meeting on 17 May 2013. Further details are provided in Section 8.1. This initial study demonstrated a current humanhealth hazard to Europe from both SO<sub>2</sub> and SO<sub>4</sub> derived from a gas emission representative of a large effusive eruption on Iceland (Figure 1.1). Previous work by Schmidt et al. (2011) amongst others had demonstrated a health hazard from SO<sub>4</sub>, but such a hazard from SO<sub>2</sub> had been less well recognised and demonstrated the need for further work to quantify the hazard from both pollutants.



Figure 1.1: Example "worst-case" hourly boundary-layer levels simulated in the initial modelling of this period for (a) sulphur dioxide (at 18:00UTC 27/06/2010) and (b) sulphate aerosol (at 03:00 01/07/2010) plotted on the UK Air Quality Index scales (see Section 5).

# **1.2 Questions to be answered**

The aim of the EEMP project was to answer the following questions for both the near-surface in the UK and a limited number of aircraft flight levels over the North Atlantic/European flight region:

- What are the typical first (fastest) arrival times for the plume of gas and/or aerosol to reach the UK?
- What is a typical maximum concentration of sulphur dioxide and sulphate aerosol that could occur?
- How long could concentrations remain elevated above defined thresholds?
- How frequently could concentrations above defined thresholds occur during a Laki-type effusive eruption?

The following questions specific to the UK land-surface were also to be answered:

- What rates of acid deposition (wet and dry) of sulphur species could be expected over the UK?
- What are the effects of these enhanced acid deposition rates on soils?

• What are the effects of enhanced ambient surface sulphur dioxide concentrations on vegetation?

Specific deliverable requirements for sulphur dioxide and sulphate aerosol were discussed with the stakeholder Departments during the project.

The intended benefits from the work were:

- A validated evidence base for the National Risk Assessment, to inform improved preparedness for this potentially national scale event.
- Consistent information for Government departments to conduct further research to determine the impacts of an effusive eruption on the aviation, health and environmental sectors.

#### 1.3 Approach

The project was conducted from October 2013 to June 2014 by the four project partners: Met Office, British Geological Survey (BGS), Centre for Ecology and Hydrology (CEH) and the University of Leeds.

To evaluate the uncertainty in the transport, deposition and therefore impact of an effusive Laki-type eruption, this work has used two atmospheric chemistry and transport models (the Met Office's NAME model and CEH's EMEP4UK model) and considered 80 meteorological evolutions spread-out over 10 years. The number and choices of simulations and models was driven by what was practical within the scope and timeframe of the work.

One of the limitations of this work, which was made clear in the planning stages, is that due to resource constraints only a single eruption scenario has been used in the modelling (this was a choice made by the funding Stakeholders). It is unlikely that this scenario will mirror the actual scenario of a future effusive eruption, both in terms of magnitude and duration. Robust investigation of the uncertainty related to the choice of scenario was also beyond the scope of the funded work.

The main objectives of the project were to provide data to the stakeholder Government Departments to be used for impact assessments. For this reason, comprehensive evaluations of all the simulations and their potential consequences were not within the scope of this work.

The work was divided into a number of Work Packages, the results from which are brought together in this report. The research outline was as follows:

- Derivation of an appropriate eruption source term based on the expert elicitation results Section 3
- Model simulations for a 10-year meteorological period Section 4
- Analysis of model results for the near-surface Section 6
- Analysis of model results for aviation relevant altitudes Section 7
- Basic assessment of the uncertainty in the modelling results related to the source term Section 8
- Assessment of the impacts on UK habitats using critical loads, critical levels and dynamic modelling Section 9
- Assessment of potential levels of halogens and hydrogen sulphide over the UK – Section 10

The conclusions from these different work strands are presented in Section 11.

A number of activities to promote and discuss aspects of the work have been conducted by the project team during the course of the project. These are outlined in

Annex A. In addition, papers and data relevant to this topic are being added to the Air Pollution Information Service (APIS) website (http://www.apis.ac.uk/), which is currently used by UK Agencies for information on air pollution and deposition effects.

# 2 Glossary

To aid non-scientific readers a comprehensive glossary has been put together to explain the technical terms that are used in this report:

*Aerosol* - suspension of solid or liquid particles in the air ranging in size from a few nanometres (nm) to around 100 micrometres ( $\mu$ m) in diameter.

Andesite - a type of volcanic rock that is common at many composite volcanoes. Andesite composition (52 to 63 wt. % SiO2) is intermediate between basalt and dacite magma. It often forms thick rubbly lava flows. However the magma usually contains moderate amounts of dissolved water and can thus produce violent explosive eruptions generating high eruption columns.

AQI – the UK Air Quality Index.

*Ash - a* fragment of volcanic rock that is less than 2 mm in mean diameter resulting from different processes of eruptive fragmentation.

*Atmospheric chemistry and transport model* – a mathematical representation of the transport and chemical reactions of gases and aerosols in the atmosphere.

*Basalt* - a type of dark-coloured volcanic rock that often forms lava flows and lowlying volcanoes. Basalt composition has less than 52 wt. % SiO<sub>2</sub>, which gives it a low viscosity and allows dissolved gases to escape from the magma. Although this type of magma often behaves in a less explosive manner than more viscous magma, basaltic magmas do erupt explosively, especially if interaction with groundwater or seawater occurs.

*Column (eruption column)* – the eruptive plume of ash and gas that rises above the *vent*.

*Critical Load* – a quantitative estimate of the pollutant deposition load below which significant harmful effects on specified elements of the environment do not occur according to present knowledge.

*Critical Level* – the gaseous concentration of a pollutant above which damage to vegetation has been observed.

*Degassing* - the process by which *volatile chemicals* that are dissolved in magma form a separate gas phase and escape from the magma. Slow degassing forms bubbles in lava flows, whereas rapid degassing can tear the magma apart explosively. Efficiency of degassing from magma before reaching the surface is one control on the explosivity of eruptions.

*Effusive eruption* - a volcanic eruption in which magma is extruded from the vent to form lava flows.

*Eruption (volcanic)* - any process on a volcano or at a volcanic vent that involves the explosive ejection of fragmented material, the effusion of molten lava, the sudden release of large quantities of volcanic gases (e.g.  $H_2O$ ,  $SO_2$ ,  $CO_2$ ), or a process by which buried regions of the volcanic systems from various depths such as the hydrothermal system are brought to the surface during edifice collapse. Eruptions are magmatic if newly solidified magma is present in the eruptive products, and are non-

magmatic if they involve only recycled rock fragments. Eruptions can occur on widely varying timescales (seconds to years).

*Eruption cloud (plume)* - a cloud of tephra and gases that forms above a volcanic vent during explosive volcanic eruptions. The vertical pillar of *tephra* and gases that forms during most explosive activity is referred to as an eruption *column*, or strong plume, and includes a momentum-dominated region and a buoyancy-dominated region. Eruption clouds may rapidly spread laterally by gravitational spreading, especially in the most energetic eruptions, and may disperse or drift thousands of kilometres downwind.

*Eruption scenario* – a description and volcanological characterisation of one eruption episode.

*Eruption source term* – a detailed scientific definition of the emitted mass, species, and the times and heights at which material is emitted from a volcano during a single *eruption scenario.* 

*Eulerian model* – a mathematical representation of motion which focuses on specific locations (three-dimensional grid cells) in space through which fluid (in this case the atmosphere and any pollution) flows as time passes.

*Explosive eruption* - a volcanic eruption in which gas bubble expansion or explosive interaction between magma and water is rapid enough to break the magma apart (i.e. fragment the magma). Explosive eruptions also occur when pressurized hydrothermal gases and superheated fluids suddenly break the host rock in a volcanic edifice.

*Fissure* – an elongated fracture on the Earth's surface that may be kilometres long through which a 'fissure eruption' releases volcanic lava, ash and gases.

*Flight level, FL* – description of altitude used by aviation, one flight level unit is equivalent to 100 ft, e.g. FL100 is 10,000 ft.

*Hydrogen chloride, HCl* - a gas that is emitted by volcanoes and is toxic at high concentrations.

*Hydrogen fluoride, HF -* a gas that is emitted by volcanoes and is toxic at high concentrations.

*Hydrogen sulphide,*  $H_2S$  - a gas that is emitted by volcanoes and is toxic at high concentrations.

*Laki-type* – A volcanic eruption scenario that is of a similar style to the 1783-1784 CE eruption of Laki, Iceland, but does not replicate that exact eruption in precise detail.

*Lagrangian model* – a mathematical representation of motion whereby the location of individual particles or fluid parcels in space and time are calculated and tracked.

*Lava* - molten rock erupted at the Earth's surface by a volcano or by an eruptive fissure, as an effusive dome or flow. When first emitted from a volcanic vent, lava is a liquid at very high temperature, typically from 700°C to 1200°C. Lava flows vary by many orders of magnitude in viscosity and this strongly influences their flow properties.

*Magma* - a mixture of molten rock (800°C to 1200°C), which also can contains suspended crystals, dissolved gases and sometimes gas bubbles. Magma forms by the melting of existing rock in the Earth's crust or mantle. Magma composition and gas content generally control the style of eruption at a volcano. In general terms, hotter, less viscous magma (e.g. basalt) allows gas to separate more efficiently, limiting the explosivity of eruption, while cooler, more viscous magma (e.g. andesite, dacite and rhyolite) is more likely to fragment violently during eruption.

*Model Source term* – the data form the *Eruption Source Term* converted in to a form that is required to initiate an *Atmospheric chemistry and transport model.* 

*Monte Carlo method* – a computational algorithm that uses repeated random sampling of input parameter distributions to generate numerical simulation results. Simulation runs are conducted many times until they adequately define the distribution and range of uncertain possible or probabilistic outcomes.

*NWP* – Numerical Weather Prediction, the use of mathematical models of the atmosphere and oceans to predict the weather using weather observations. These models are used to generate meteorological data, which is needed as an input to the NAME and EMEP4UK models.

Period – used to refer to each separate 6-week eruption that has been modelled.

*Stratosphere* - The region of the atmosphere between the *troposphere* and ~50 km (i.e. base of the mesosphere), in which the temperature generally increases with height. Material in the stratosphere does not readily mix into the troposphere.

Sulphur dioxide,  $SO_2$  - a volcanic gas and a product of combustion of sulphur containing materials that is toxic at high concentrations.

Sulphate aerosol –the product of oxidation of SO<sub>2</sub> gas to SO<sub>4</sub><sup>2-</sup>; it occurs as liquid or solid particulate matter in the atmosphere. The composition of sulphate aerosol will vary as the volcano plume interacts with the atmosphere from acidic sulphate (H<sub>2</sub>SO<sub>4</sub>) to partly or fully neutralised with ammonium, (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>. In this work sulphate aerosol with sizes of up to 2.5 micrometers (often referred to as PM<sub>2.5</sub>) is considered.

*Stochastic model* – a tool using probability distributions to estimate quantitatively potential outcomes by allowing for random variations in one or more inputs over time or space. In the present case, uncertainty distributions for input parameters in a future *Laki-style* eruption were defined by expert elicitation to represent uncertainties on key variables and factors.

*Tephra* - any type of solid material erupted from a volcano, regardless of size, shape, composition or method of formation.

*Tropopause* – upper limit of the *troposphere* and the boundary between the *troposphere* and the *stratosphere*. This occurs at a height of between 10-17 km. Above Iceland the tropopause is at approximately 10 km.

*Troposphere* - the lowermost portion of the atmosphere from the Earth's surface to the *tropopause*. The troposphere is the portion of the atmosphere where most of the clouds and weather occur.

*Vent* – the surface opening from which volcanic material is erupted. Vents may be either circular structures (i.e. craters) or elongate *fissures* or fractures, or small cracks in the ground.

*Volatile* - a dissolved component in a magma at high pressure and temperature, which forms a separate gas phase at lower pressure or temperatures. The most common volatile in magma is water, followed by carbon dioxide and sulphur dioxide; other volcanic volatiles include elements or compounds such as HCI or HF that form a gas at relatively low pressures and temperatures.

*Volcanic event -* any occurrence, or sequence of phenomena, associated with volcanoes that may give rise to volcanic hazards.

*Volcano* - a naturally occurring *vent* at the Earth's surface through which magma, solid rock, and associated gas and water can erupt. A volcano is also the edifice that is built by the explosive or effusive accumulation of these products over time.

*VSD* - "Very Simple Dynamic" model; a model that is used to investigate the long term effects of acid (sulphur and nitrogen) deposition on habitats.

# **3** Definition of the Eruption Source Term

It is essential to have sensible 'source terms' for any model simulations conducted, otherwise the model outputs will have minimal value or meaning. Good historical observations in Iceland in 1783-84 and excellent geological investigation mean there is a sound scientific basis for understanding the Laki eruption (e.g. Thordarson and Self 1993; 2003), which has formed the basis for assessments such as Schmidt et al. (2011). Nevertheless, for detailed modelling of possible future scenarios there is a need to capture the likely range of uncertainty for some of the eruption parameters.

In 2012 the Civil Contingencies Secretariat requested that the source characteristics of a future Laki-type eruption scenario be considered and documented to facilitate modelling of the potential future impacts on the UK. It was proposed that an expert elicitation would be the best way to establish preliminary values for source characteristics and their uncertainties, and therefore to bring together a multidisciplinary group of experts. The task was commissioned in April 2012 and scientists were asked to consider a fissure eruption of similar duration to Laki (8 months) and to give probability distributions for parameters such as plume height, magma production rates and SO<sub>2</sub> content. It is unknown what a future eruption of this type will look like so an expert elicitation is a means for giving expression to the range of possibilities based on current knowledge, by capturing and formally combining the group's informed judgements. Scientists were also asked to give assessments of some source characteristics not available in the literature; the vertical distribution of SO<sub>2</sub> in the eruptive column is one topic that proved challenging. The task was commissioned in April 2012 and the outcomes are presented in Loughlin et al. (2013) along with a description of the eruption and its impacts based on published research.

The effusive eruption modelling project which began in 2013 set slightly different constraints on the source parameters required. Given the time constraints of the project, the Met Office suggested that the full 8 month eruption scenario should not be run in its entirety but that a smaller 'subset' of parameters - which could be repeated more times with different meteorology scenarios - would be more appropriate. This was a case of optimising the modelling time available by using as wide a variety of meteorology scenarios as possible. The project team agreed that a 6-week eruptive sequence would be appropriate given the analysis time available and our understanding of the Laki eruption as described below. In order to 'reset' after each scenario, a week of no emissions was required (the 6<sup>th</sup> week) so a stochastic SO<sub>2</sub> source model for a 5 week eruption period was necessary.

# 3.1 Laki eruption

The Laki eruption dynamics are well characterised in several papers (e.g. Thordarson and Self, 1993; 2003). The eruption was intense for the first three months, with a decline in intensity thereafter (Fig. 3.1). The first five weeks of the eruption represent the most intense period overall. As can be seen from the figures, in the first five weeks there were five eruption episodes, each starting with an explosive phase and each episode representing the opening of a new fissure. Magma discharge rate was highest during the explosive onset of each episode. More than 60% of the total SO<sub>2</sub> mass was released in the first 5 weeks according to Thordarson and Self (2003).



Figure 3.1: (a) Time series showing each fissure opening episode, explosive phases and relative magma discharge rate. (b) The  $SO_2$  mass released by individual episodes, lava degassing and cumulatively through the eruption (1Mt = 1Tg). Thordarson and Self (2003).

# 3.2 Methodology

We have developed a stochastic model for Laki  $SO_2$  production which generates multiple scenario-based column  $SO_2$  release profiles for input to the atmospheric chemistry and transport models. Each model output comprises a 6-week duration time series with 24-hour time-steps enumerating average and peak effective column heights at each step, with corresponding height-dependent magma flux rates (Figs. 3.2 and 3.3). From these data, the cumulative amount of  $SO_2$  released can be totalled in Tg (Fig 3.4). The model can be sequenced into a series of discrete explosive phases in the early stages of the eruption, producing gas throughout the height of the column; these highly explosive phases are intercalated with lower level flux outputs, so that at no time in the first 5-weeks is gas production zero.

The stochastic model also incorporates a rudimentary  $SO_2$  height level-mass release determination, which is more realistic than assuming a steady emission with height. It was necessary to create this due to considerable uncertainty among experts in 2012.

Based on discussions during the expert elicitation, we have created a conceptual approach as follows:

Column height is partitioned into 5 levels:

- 1. maximum height
- 2. midpoint between maximum height and average height
- 3. average height level
- 4. 8/10ths of average height
- 5. one-third of average height

with total available  $SO_2$  in each column (i.e. the total scenario  $SO_2$  in Tg is derived from scenario magma volume erupted, divided pro rata into N explosions steps and M effusive phase steps in relation to column max height) for each time-step iteration; these are portioned out in the column at the defined five heights in packets of 7%; 20%; 50%; 20%; 3% respectively. This conceptual height release scheme looks, to a first approximation, like a 'typical' column/plume – giving off most material at uppercentral levels but with variations in relation to column height in the individual case (Table 3.1).

With this basic model, we can generate as many time outcomes as needed by using Monte Carlo re-sampling of defined uncertainty distributions (from the expert elicitation), producing multiple source terms with which we could explore sensitivity and variability using dispersion modelling.

Alternative methods to the Monte Carlo ensemble approach are possible, but this approach was agreed by the project team.

#### 3.2.1 Assumptions

There are significant uncertainties associated with any consideration of the source term of a future eruption. In order to create a stochastic model in the presence of such uncertainties we have made some basic assumptions, as follows:

- 1. At the source, SO<sub>2</sub> and magma eruption rate are coupled, i.e. higher magma eruption rates correspond to larger emissions of SO<sub>2</sub>. However, we know that their behaviour is largely decoupled and therefore larger SO<sub>2</sub> mass may be released at any given time.
- 2. Vertical distribution of  $SO_2$  in a column mimics the distribution of ash in the column. In the absence of knowledge to the contrary, this is a typical working assumption about eruptions. We do know that  $SO_2$  would, additionally, be released at the vents *and* from erupting/erupted lavas so it would be present at low and high altitudes in the atmosphere.
- 3. In this work we consider only SO<sub>2</sub>. These types of eruptions also produce volcanic ash (especially if erupting through groundwater, surface water or snow/ice) and other gases and particulates including trace metals.

#### 3.3 The Scenario

Databases of multiple stochastic model runs have been created, each holding 24hour source terms for a Laki-type eruption extending over a period of 6 weeks. These include information on plume height and  $SO_2$  mass erupted for a height profile of five layers for each time-period, which allows a more realistic emission variation with height.

Databases of 6-hour source terms were also prepared but it was decided that to prevent false interpretation of temporal detail generated in the source term and reduce computational costs only the 24-hour sources would be used.

One scenario, representing a "precautionary worst case", was suggested and agreed for the modelling in this project. This scenario has sustained elevated tropospheric plume height (6+ km) for the first two weeks and for much of the rest of the period. An assumption made here is that  $SO_2$  in tropospheric plumes at altitudes of 6+ km are most likely to be carried large distances. The overall  $SO_2$  emission derived from the stochastic model for the selected 5-week scenario was 72 Tg, which is similar to the actual total  $SO_2$  emission (vent + lavas) for the Laki eruption during a similar time period (69 Tg, Thordarsson and Self, 2003). In this scenario, 47 Tg of the emitted  $SO_2$  goes into the eruptive columns above the vents and the remainder is emitted from the lava flows. Only the explosive eruptive columns have been modelled in this work.

Multiple alternative scenarios could be used for further studies investigating source term uncertainty and related topics.



Figure 3.2: Example of a single 6-week eruption scenario, which corresponds to the scenario used in the modelling. Heights are in km above the volcanic vent.

Magma Flux Rate



Figure 3.3: Corresponding magma flux rate for the same scenario as Figure 3.2.

Cum SO2 output





	Height profile [5 levels in km]				SO <sub>2</sub> mass Tg at height levels					
Timestep	1	2	3	4	5	1	2	3	4	5
1	11.5	9.2	6.9	5.5	2.1	0.115	0.330	0.824	0.330	0.049
2	14.6	12.8	10.9	8.7	3.3	0.147	0.420	1.050	0.420	0.063
3	10.9	9.7	8.5	6.8	2.6	0.110	0.314	0.785	0.314	0.047
4	15.6	14.4	13.3	10.6	4.0	0.157	0.450	1.124	0.450	0.067
5	13.7	12.5	11.3	9.1	3.4	0.138	0.394	0.986	0.394	0.059

Table 3.1: The SO<sub>2</sub> mass vertical distribution outputs for the first five days of the scenario used in the modelling. Heights are in km above the volcanic vent.

## 3.4 Conversion of the Scenario into a model source term

In order to model a Laki-type eruption, atmospheric chemistry and transport models require a "model source term" that describes the height, duration and mass of material emitted. The "eruption source term" provides a vertical distribution for the  $SO_2$  emission, which is defined by five spot-heights in an emission profile (Fig 3.5(a)). The eruption source term contains a separate vertical profile for every 24-hour emission in the 5-week eruption. Atmospheric models can release emissions at spot heights or over vertical ranges. In the Laki-type case, if the models were to only emit at the five discrete spot heights then the true distribution of  $SO_2$  with height would be incorrectly represented and the modelling results would be biased by the prevailing winds at the five spot heights. Instead, the models need to release the emissions over representative vertical layers that provide a match to the original vertical distribution (Fig 3.5(b)).

There are a number of ways to approach the choice of vertical layers, but a fundamental requirement in each case is to define a top and bottom height for each layer. To define five layers, six heights are required. As can be seen from Figure 3.5(b) with the current distribution we do not know at what heights the emission is zero. These "zero-heights" are needed to define the maximum and minimum heights of the emission. To enable the derivation of the model source term the following, admittedly arbitrary, assumptions have been made:

- The emission is zero at the volcanic vent. The vent height is taken to be at 600 m above sea level, which is typical for the Laki region in Iceland.
- The emission is zero 300 m above the maximum height provided in the source term.

The resulting distribution (interpolated between the spot heights for graphical representation of the shape of the distribution) is depicted in Figure 3.6.



Figure 3.5: (a) The five spot heights for one 24-hour emission. (b) An example emission distribution created from the five spot heights (Note: the smoothed line demonstrates the vertical emission profile, but cannot be used to determine the mass distribution between the discrete spot heights). Height is in km above sea level.



Figure 3.6: The emission distribution depicted in Figure 3.5, but with top and bottom heights included as described in the text.

Taking the five provided spot height values and using the zero-emission heights as the top and bottom of the distribution gives the five numbered layers shown in Figure 3.7. Overlaying these layers on the original distribution (Fig 3.7) demonstrates that this has the effect of lowering the height of the emission in the atmosphere compared to the original distribution. For example, the maximum emission occurs between approximately 6 km and 10 km in the original distribution, but the layer 3 height range puts this between approximately 6 km and 7.5 km. There is also considerably more material emitted at the lower levels (4 and 5) than the original distribution would suggest.

A better alternative is to use the mid-points between the original heights as the top and bottom heights of the model layers. This gives the distribution shown in Figure 3.8, which gives a better fit to the original distribution and maintains the height range of the maximum emission. For this reason, this "mid-point" distribution has been used to define the emission layers for the modelling. In this mid-point distribution, the mass at each spot height is uniformly emitted between the top and bottom heights of the corresponding vertical layer. For example, in Figure 3.5 the spot height 7.45 km (asl) has an SO<sub>2</sub> emission mass of 0.824 Mt, with no emissions between this height and next spot height of 9.76 km (asl) or the lower spot height. In the mid-point model distribution in Figure 3.8, the 0.824 Mt is uniformly released between 6.77 km (asl) and 8.61 km (asl) and so on, and there are no heights with zero emission between the original spot heights.



Figure 3.7: The layers achieved (green) if the original distribution heights (blue diamonds) are used to define the top height of each layer.



Figure 3.8: The five layers achieved (orange) if the mid-points of the original distribution (blue) are used to define the top and bottom heights of each layer.

#### 3.4.1 Lower emission heights

The eruption sequence contains 5-weeks (35 days) of differing eruption heights. All of these are described using five spot heights, but for some of the lower eruption heights (< 8 km asl), the top three heights have the same height value but are still provided with separate emission amounts (Fig 3.9). The discretised source for those phases has been constructed using a slightly different approach whereby the five heights are reduced to a 3-layer emission profile by combining the emissions of the top three heights into one layer. Using the same assumptions as above for defining the top and bottom emission heights and the mid-point approach gives the distribution depicted in Figure 3.10. This three-layer distribution applies to 10 of the eruption days.



Figure 3.9: An example emission distribution for a "lower" emission height phase.



Figure 3.10: A three-layer emission profile for use when the eruption column height is lower than ~8 km above sea level.

## 3.5 Full eruption source term sequence

Figure 3.11 shows the full range of emissions profiles for the 35 days with emissions in the 6 week sequence. As outlined a vent height of 600 m has been assumed, so the profiles are plotted above sea level. Figure 3.11 shows the emissions dataset that has been used in the modelling.

This 6-week eruption scenario has then been modelled eighty, non-overlapping, times over 10 years of meteorology from 2003-2012. Due to limitations with the EMEP4UK model, no emission periods could be run over two years (i.e. crossing from December into January), so each year contains 8 discrete simulations. This means that the last week in December is not represented in any of the situations, but this is unlikely to introduce a bias in the meteorology beyond the existing sampling limitations. Start dates for each of the simulation periods are given in Table 3.2 and Figure 3.12 pictorially demonstrates how the emissions are repeated within one year.

01/01/2003	16/01/2005	01/01/2007	16/01/2009	01/01/2011
12/02/2003	27/02/2005	12/02/2007	27/02/2009	12/02/2011
26/03/2003	10/04/2005	26/03/2007	10/04/2009	26/03/2011
07/05/2003	22/05/2005	07/05/2007	22/05/2009	07/05/2011
18/06/2003	03/07/2005	18/06/2007	03/07/2009	18/06/2011
30/07/2003	14/08/2005	30/07/2007	14/08/2009	30/07/2011
10/09/2003	25/09/2005	10/09/2007	25/09/2009	10/09/2011
22/10/2003	06/11/2005	22/10/2007	06/11/2009	22/10/2011
08/01/2004	24/01/2006	08/01/2008	24/01/2010	08/01/2012
19/02/2004	07/03/2006	19/02/2008	07/03/2010	19/02/2012
01/04/2004	18/04/2006	01/04/2008	18/04/2010	01/04/2012
13/05/2004	30/05/2006	13/05/2008	30/05/2010	13/05/2012
24/06/2004	11/07/2006	24/06/2008	11/07/2010	24/06/2012
05/08/2004	22/08/2006	05/08/2008	22/08/2010	05/08/2012
16/09/2004	03/10/2006	16/09/2008	03/10/2010	16/09/2012
28/10/2004	14/11/2006	28/10/2008	14/11/2010	28/10/2012



Table 3.2: The start dates of each of the eighty 6-week periods used in this work.

Figure 3.12: Example showing how the model source term is repeated in Year 1.



Figure 3.11: Every daily emissions profile in the 6 week sequence. All 35 profiles are depicted, although the legend only has space for 30.

## 3.6 Characteristics of the modelling source term

- The total emission of  $SO_2$  over the 5-weeks is 47.309 Mt. This is followed by 1 week of zero emission
- The maximum daily emission is 2.248 Mt
- The maximum hourly emission rate is 0.094 Mt/hour
- The maximum height of emission is 16.4 km above sea level
- On the maximum emission day (day 4), the bulk of material is emitted into the stratosphere.
- The maximum daily emission in any height layer is 1.124 Mt
- Making the crude assumption that any layer wholly above 10 km asl is in the stratosphere (a reasonable assumption over Iceland) then 11.23 MT (~24%) of SO<sub>2</sub> is emitted into the stratosphere and 36.079 MT (~76%) is emitted into the troposphere. The temporal variation in this split is shown in Figure 3.13.



Figure 3.13: (a) The mass of SO<sub>2</sub> released into the troposphere and stratosphere for each 24hour emission, assuming that over Iceland the tropopause is at 10 km. (b) The maximum emission height on each day.

# 4 Model descriptions

Two atmospheric chemistry and transport models (ACTM) have been used: the Met Office's NAME model and the Centre for Ecology and Hydrology's EMEP4UK model. This section provides a brief introduction to the models and a summary of how they have been set up to provide the required data for this work.

# 4.1 The EMEP4UK model

The EMEP4UK model is a nested regional Eulerian model based on version v4.3 of the main EMEP model (Simpson et al., 2012). A detailed description of the EMEP4UK model framework and setup are given in Vieno et al. (2010) and Vieno et al. (2014).

### 4.1.1 EMEP4UK sulphur chemistry

The EmChem09 chemical mechanism has been used for this work, which contains 72 species and 137 reactions. The sulphur and nitrogen chemistry are summarised in Table 4.1. Cloud scavenging and wet and dry deposition are applied to all species.

Sulphate chemistry	Nitric acid chemistry
OH + SO2 ⇒HO2 + SO4	NO + NO3⇒ NO2 + NO2
SO2 + H2O2 ⇒ SO4	NO + HO2⇒ NO2 + OH
SO2 + O3 ⇒SO4	NO2 + NO3⇒ NO + NO2
SO2 + Fe⇒SO4	OH + HO2⇒ H2O + O2
	OH + H2O2⇒ HO2 + H2O
Aerosol equilibrium:	OH + H2⇒ HO2 + H2O
$SO_4^{2-}HNO_3-NO_3^{-}-NH_3-NH_4^{+}$	OH+HNO3 OH + HNO3⇒ NO3 + H2O
	HO2+HO2 HO2 + HO2⇒ H2O2
	$OH + HONO \Rightarrow NO2$
	N2O5 ⇒2. HNO3
	k <sub>aero</sub> HNO3 ⇒NO3 c
	NO+OP OP + NO + M $\Rightarrow$ NO2
	NO2 + NO3⇒ N2O5
	N2O5 ⇒NO2 + NO3
	NO2+OH NO2 + OH + M ⇒HNO3
	OH+NO OH + NO⇒ HONO

Table 4.1: Summary of sulphur and nitrate chemistry in EMEP4UK.

#### 4.1.2 Anthropogenic emissions

EMEP4UK includes anthropogenic and biogenic emissions as standard. Anthropogenic emissions are included for NOx,  $NH_3$ ,  $SO_2$ , primary  $PM_{2.5}$ , primary PMcoarse, CO, and non-methane volatile organic compounds (NMVOC). These are derived from the following sources:

- For the UK, emissions values are taken from the National Atmospheric Emission Inventory (NAEI, http://naei.defra.gov.uk) at 1 km<sup>2</sup> resolution and aggregated to 5 km x 5 km resolution
- For the rest of the domain, the model uses the EMEP 50 km x 50 km resolution emission estimates provided by the Centre for Emission Inventories and Projections (CEIP, http://www.ceip.at/). These include emissions for Etna.
- Emissions estimates for international shipping (ENTEC, 2010) are aggregated to 5 km x 5 km for those emissions within the inner domain.
- The land-based gridded emissions are distributed vertically according to a default distribution based upon the SNAP codes (Simpson et al., 2012). The emissions have a temporal resolution of hours.

# 4.2 The NAME Model

NAME is a Lagrangian particle model in which large numbers of model particles are released and tracked through the model atmosphere. Each model particle represents a certain mass of the released material and the transport and dispersion of this material is governed by input meteorological data. The motion of the particles also has a random component added to represent the effects of atmospheric turbulence. Pollutants can be removed from the model atmosphere by several processes including fall out due to gravity, impaction with the surface, washout where the pollutant is 'swept out' by falling precipitation, and "rainout" where the pollutant is absorbed directly into cloud droplets as they form. For further details on the model physics see Jones et al. (2007).

NAME is used to provide the modelling service for the London Volcanic Ash Advisory Centre (VAAC) and other operational forecasts e.g., for radiological and chemical hazards, issued by the Met Office Hazard Centre. It also includes the necessary chemical reactions for simulating volcanic SO<sub>2</sub> and SO<sub>4</sub> that are of interest in this work. This chemistry capability in NAME was originally developed for routine Air Quality forecasting purposes. However, since 2011 work has been undertaken by the Met Office and the University of Leeds to evaluate this capability in relation to volcanic emissions (Heard et al., 2012; Schmidt et al., 2014). This work has verified the NAME output against satellite and airborne observations of SO<sub>2</sub> and SO<sub>4</sub> from a number of volcanic eruptions and shown that the model is suitable for this application.

The chemistry calculations are performed on a fixed three-dimensional Eulerian chemistry grid. The initial species concentrations in a chemistry grid-box are obtained by summing the contributions from all particles occupying that box at the given time. Following completion of the chemistry calculations, the updated mass of each species in the chemistry box is reassigned back to these particles: primary pollutants being redistributed according to the relative proportion of the original contributions, and with secondary species (in this case sulphate) being distributed among particles carrying the appropriate primary species in proportion with the original amount of primary pollutants (Jones et al., 2007). The chemistry scheme is described in more detail by Redington et al. (2009) and Heard et al. (2012).

## 4.2.1 NAME sulphur chemistry

 $SO_2$  is converted to sulphate in the atmosphere by reactions involving OH, hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) and ozone (O<sub>3</sub>). The gas phase reaction involving sulphate is:

$$OH + SO_2 + M \rightarrow HSO_3 + M$$

where M denotes ambient air. This reaction is dependent on temperature and pressure, and controls the rate of sulphate production as  $HSO_3$  is rapidly oxidized to  $H_2SO_4$ . This  $SO_2$  gas phase reaction is simulated throughout the atmosphere in NAME and dominates in the stratosphere where there is little cloud present.

Aqueous phase reactions take place if the NAME grid box contains a non-zero cloud fraction and cloud water, and these reactions dominate sulphate production in cloudy conditions. There are two pathways for the oxidation of SO<sub>2</sub> in the aqueous phase, involving either  $H_2O_2$  or  $O_3$  in solution. The reaction with  $H_2O_2$  is very rapid. The reaction with  $O_3$  is limited by the pH – as more  $H_2SO_4$  is produced, the acidity of the cloud increases and the reaction rate slows.

The gas to liquid phase equilibria, in which SO<sub>2</sub> dissolves and dissociates, are:

 $H_2O + SO_2 \rightleftharpoons H^+ + HSO_3^-$ 

 $HSO_3^- \rightleftharpoons H^+ + SO_3^{-2-}$ 

The aqueous phase reactions relevant to the production of sulphate are:  $HSO_2^- + H_2O_2 \rightarrow SO_2^{2-} + H^+ + H_2O_2$ 

$$\begin{split} ISO_{3}^{-} &+ H_{2}O_{2} \rightarrow SO_{4}^{2^{-}} + H^{+} + H_{2}O \\ HSO_{3}^{-} &+ O_{3} \rightarrow SO_{4}^{2^{-}} + H^{+} + O_{2} \\ SO_{3}^{2^{-}} &+ O_{3} \rightarrow SO_{4}^{2^{-}} + O_{2} \end{split}$$

The concentrations of the photo-oxidant species hydroxyl (OH) and hydroperoxyl radical (HO<sub>2</sub>) are modelled explicitly within NAME. Ozone (O<sub>3</sub>) and hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) are also calculated within the model, being initialised at the start of a model run using values taken from background fields provided by the STOCHEM model (Collins et al., 1997). NAME has been run using the chemistry scheme option of "ozone on background fields", which means that NAME does not require chemical lateral boundary conditions for these runs

In a simulation using full anthropogenic emissions, the available model aerosol sulphate would be rapidly combined with ammonia to form ammonium sulphate near the surface.

Further details on the reactions and equilibrium included in the NAME chemistry scheme are given in Redington et al (2001).

#### 4.2.2 Number of model particles

The number of model particles controls statistical 'noise' in the results (more particles result in less noise) and affects the amount of time each simulation takes (more particles result in longer run times). Here the number of model particles emitted per source is proportional to the mass of the  $SO_2$  emission. For the smallest release of 0.01Tg, this equates to 2222 particles over 24 hours, which is equivalent to 92 particles per hour. This is less than a typical modelling minimum, but calculations show that in all cases this provides more than 10 particles per hour per 100m of height in the source profile. This is deemed to be sufficient to represent the source at a suitable signal to noise ratio.

## 4.3 Modelling set-up

#### 4.3.1 Volcanic emissions

Volcanic emissions have been used as defined in Section 3.

- The eighty separate emissions periods have been simulated in each model.
- The location of the volcano is taken to be at N64.064 and W18.22.
- In NAME each source is represented by a 100 m diameter cylinder extending from above the volcano summit to the maximum height of the eruption column. SO<sub>2</sub> is emitted in three or five layers within the cylinder as defined by the model source term.
- In EMEP4UK, the volcanic emissions are injected in the appropriate model level at a horizontal resolution of 50 km<sup>2</sup>; emissions with a height above the model vertical domain are injected in the highest model level.
- In EMEP4UK anthropogenic emissions of other species were also included, as detailed above. In NAME only volcanic SO<sub>2</sub> was emitted, no other biogenic or anthropogenic emissions were included.

#### 4.3.2 Domain and Resolution

• NAME typically outputs on a regular latitude-longitude grid, whereas the EMEP4UK grid is on a rotated pole (Fig 4.1). The closest approximation to



the EMEP4UK grid in a regular lat-lon framework is a domain of 25N to 85N and 60W to 60S and this is what has been used in NAME.

Figure 4.1: (a) EMEP4UK domains and topography representation. The red box indicates the nested 5 km<sup>2</sup> model domain. (b) The NAME dispersion and output domain used in this work.

- The horizontal resolution required was 25 km x 25 km.
  - This has been approximated in NAME by outputting on a regular 0.25 degree by 0.25 degree grid over the whole domain.
  - For EMEP4UK the model horizontal resolution scales down from 50 km x 50 km in the main EMEP 'Greater European' domain (which includes Iceland) to 5 km x 5 km for the domain covering the British Isles as shown in Figure 4.1.
- The vertical resolution used is model specific.

- For NAME's three-dimensional chemistry grid a resolution of 200 m below 1 km and 500 m above 1 km agl was chosen.
- For EMEP4UK the model layers are based on a pressure coordinate system and so the heights listed for each layer will vary at each timestep as the surface pressure changes. An example of the vertical structure of the model is shown in Figure 4.2. The top of the model domain is at approximately 16 km above sea level.



Figure 4.2: The EMEP4UK vertical layer structure.

- Output for the near-surface has been taken as:
  - An average over a depth of 0-200 m above ground level for NAME.
  - The lowest model layer, which is approximately ~90m (model layer 0), for EMEP4UK.
- Output is also required for the heights in Table 4.2, which are relevant to the aviation sector.
  - To obtain a full vertical profile in NAME the standard VAAC "thin" vertical levels, which are layers of 2500 ft depth, have been used. For the specific layers requested for the aviation sector analysis the nearest of these layers has then been used as shown in Table 4.2.
  - For EMEP4UK the model layer bounding the approximate height has been used.
| Description                       | <b>Required Height</b> | NAME layer range | EMEP4UK layer                       |
|-----------------------------------|------------------------|------------------|-------------------------------------|
| Standard cruising altitude        | 37,000 ft = FL370      | FL350-FL375      | model layer 16<br>~10300 - ~11490 m |
| Reasonable cruising altitude      | 30,000 ft = FL300      | FL275-FL300      | model layer 15<br>~9050 - ~10350 m  |
| Lowest on route level             | 24,000 ft = FL240      | FL225-FL250      | model layer 13<br>~6200 - ~7620 m   |
| General aviation<br>aircraft      | 15,000 ft = FL150      | FL125-FL150      | model layer 11<br>~4040 - ~5000 m   |
| Holding stacks                    | 7000 ft = FL070        | FL050-FL075      | model layer 8<br>~1900 - ~2500 m    |
| Low-level flying and recreational | 1000 ft =FL010         | FL000-FL025      | model layer 2<br>~180 - ~320 m      |

 Table 4.2: Heights required by the Department for Transport and the associated model output levels.

#### 4.3.3 Meteorological data used

The EMEP4UK model is driven by the Weather Research Forecast (WRF) model version 3.1.1 (www.wrf-model.org). The boundary conditions for the inner domain are derived from the results of the European domain in a one-way nested setup. The EMEP4UK model uses a yearly boundary condition derived from observation for SIA at the edge of the European domain adjusted for each year as describe in Simpson et al. (2012).

NWP meteorology from the Global forecast set-up of the Met Office's Unified Model has been used for all years as input to NAME. As this is an operational forecast model, its resolution and physics have improved over the years considered in this study. Table 4.3 outlines the data that has been used in this work.

Periods	Resolution	Top height
01/01/2003-06/12/2005	60 km	19 km
06/12/2005-31/12/2008	40 km	19 km
01/01/2009-10/11/2009	40 km	19 km
10/11/2009-09/03/2010	40 km	19 km
09/03/2010-31/12/2012	25 km	29 km

Table 4.3: Summary of the Met Office Unified Model NWP data used as input to NAME.

#### 4.3.4 Run options and output quantities

- Both models have been run with wet and dry deposition turned on and deposition amounts have been output..
- Air concentrations with hourly and 24-hour output temporal resolution have been produced.
  - For the 24-hour data, a daily 24-hour mean output has been produced. This enables the SO<sub>4</sub> outputs to be compared to the Air Quality index. A 24-hour rolling mean or any other temporally averaged (t > 1 hr) output could be obtained by post-processing the hourly data.

#### 4.4 Assumptions and Limitations

Decisions on how to represent a given event, the processes captured and the exact form of the output from a modelling study are dependent on the specific model, the modellers experience and judgment, and the time available to perform a given study. In a multi-model study, such as here, these differences often add to the robustness of the findings by enabling the exploration of the sensitivity of the solutions to such choices. However certain differences can appear confusing or even concerning to non-modellers. In this study there are certain items worth noting in relation to this:

- In the results presented here, near-surface is taken to be the average over a 0-200 m layer in NAME and the ~90 m model layer in EMEP4UK. At the distance of the UK from the volcano, such a difference in depth has a negligible impact as both layers are within the planetary boundary layer where the atmosphere is well mixed.
- For flight levels, the NAME model outputs are the average concentration in the 2500 ft flight level layer given in Table 4.2. This volume average is necessary due to the Lagrangian modelling framework and to ensure an adequate signal-to-noise ratio.
- The flight levels for the EMEP4UK model are extracted from the relevant model layer. The EMEP4UK model uses 20 vertical layers, with terrain following coordinates, and resolution increasing towards the surface. The layers are based on pressure so the exact height of each layer in metres changes. The model extends from the surface up to 100 hPa (~16 km).
- EMEP4UK results include anthropogenic emissions and other non-scenario low-level volcanic emissions from Etna of both SO<sub>2</sub> and SO<sub>4</sub>. NAME results do not.
- The release dimensions of the two models are different: EMEP4UK is limited to releasing into a grid box volume, whereas NAME releases from a 100m diameter cylinder above the volcano.
- The two models use different sources of NWP meteorology.
- The physics and chemistry schemes are different in the two models, although both have been used in the Defra model intercomparison and show similarity for air quality assessments.

There have had to be a number of limitations in the way that the model runs and analysis have been conducted in order to fit within the resource available to and time frame of this project.

- The dispersion domain is fixed at the sizes shown in Figure 4.1. No recirculation of any material that leaves the domain is allowed. However it is considered very unlikely that this would impact on the final results at a level of practical significance.
- While the number of scenarios considered here is considerable, it is still
  insufficient to capture the entire range of variability in the numerous aspects
  of such an event and, in particular, only one eruption scenario has been
  considered. Whilst this work presents a good assessment of the potential
  impact, it is not exhaustive and may potentially under- or over-estimate the
  likely impact of such an event.
- This report focuses on results for the UK only.
- Outputs from the two models are presented side-by-side. No attempt has been made to combine the results into one ensemble.
- The results presented do not account for the uncertainty that is inherent in this study. Section 8 provides an assessment of the range of uncertainty that should be taken into account when analysing these results.
- No attempt is made here to look at the consequences of these results or their application to impact assessment.

# 5 Air Quality Scales and Species

The modelled concentrations of SO<sub>2</sub> and SO<sub>4</sub> in Sections 6 and 7 are presented on the Daily Air Quality Index scales that have been extended by Public Health England (PHE) for the purposes of this work (Tables 5.1 and 5.2). Table 5.1 shows the index scales for 15-minute mean concentrations of SO<sub>2</sub> with additional index 11, 'Hazardous' and index 12 'Very Hazardous', based on 24-hour means, and index 13 'Very Hazardous' based on 10-60 minute means. Table 2 shows the index scales for PM<sub>2.5</sub> based on 24-hour mean concentrations (for historical data this is normally the daily mean concentration, whereas for air quality assessment this is generally the latest 24 hour running mean for the current day). Again three further index values have been added: index 11, 'Hazardous', index 12 'Very Hazardous' and index 13 'Very Hazardous'. The World Health Organisation 24-hour mean guidelines have also been considered, which are 20  $\mu$ g/m<sup>3</sup> for SO<sub>2</sub> and 25  $\mu$ g/m<sup>3</sup> for SO<sub>4</sub>. It should be noted that the use of these index bands obscures some of the finer level detail in the geographical distribution that is present in the results, particularly in the lower concentration bands.

Data are presented as "hourly" values, which are derived from 1-hour mean air concentrations, and "daily" values, which are derived from 24-hour mean air concentrations. All of the statistics and maps are derived from the eighty 6-week scenarios (also referred to as "Periods"), so the daily means are derived from 3360 (80x6x7) 24-hour means and the hourly values from 80640 (80x6x7x24) 1-hour means. Following discussion with PHE and the Department for Health, the Air Quality Index (AQI) values have been applied to both the hourly and daily averages, but the appropriateness of each should be considered when interpreting the results.

At the distance of the UK from Iceland (1000+ km) any plume will be reasonably well mixed by the time it reaches the surface, so it is expected that the hourly concentrations will be representative of the 15-minute averages specified for  $SO_2$  in the index.

All concentration data are given at ambient temperatures and pressures in units of  $\mu g/m^3$ . For the near-surface these can be converted to ppb using standard atmospheric pressure and temperature values.

In order to understand the significance of the modelled outputs in this study, it is useful to put them into context by briefly considering the current levels of  $SO_2$  and  $PM_{2.5}$  over the UK. The information and figures below are extracted from "Air Pollution in the UK 2012, Department for Environment Food and Rural Affairs, September 2013".

Man-made sulphur dioxide (SO<sub>2</sub>) is an acid gas formed when fuels containing sulphur impurities are burned. The largest UK source is currently power generation. Other important sources include industry, commercial fuel use, and residential fuel use in some areas. SO<sub>2</sub> is a respiratory irritant that can cause constriction of the airways and people with asthma are considered to be particularly sensitive. Health effects can occur very rapidly, making short-term exposure to peak concentrations important. Annual mean concentrations are typically less than 5  $\mu$ g/m<sup>3</sup> except at sites in industrial locations or in residential areas with high use of solid fuel for heating. Figure 5.1(a) shows the 99.73rd percentile of 1-hour mean SO<sub>2</sub> concentration in 2012 and Figure 5.1(b) shows the 99.18th percentile of 24-hour mean SO<sub>2</sub> concentration in 2012.



Figure 5.1: (a) 99.73rd percentile of 1-hour mean SO<sub>2</sub> concentration, 2012 ( $\mu$ g/m<sup>3</sup>). (b) 99.18th percentile of 24- hour mean SO<sub>2</sub> concentration, 2012 ( $\mu$ g/m<sup>3</sup>)

 $PM_{2.5}$  can be primary (emitted directly to the atmosphere) or secondary (formed by the chemical reaction of other pollutants in the air such as  $SO_2$  or  $NO_2$ ). The main source is combustion, e.g. vehicles and power stations. Sulphate aerosol ( $SO_4$ ) is one of several secondary components of  $PM_{2.5}$ . Fine particulate matter can penetrate deep into the lungs and research in recent years has strengthened the evidence that both short-term and long-term exposure to  $PM_{2.5}$  are linked with a range of health outcomes including (but not restricted to) respiratory and cardiovascular effects. Annual mean urban  $PM_{2.5}$  concentrations in the UK are typically in the low teens of  $\mu g/m^3$  but exceed 20  $\mu g/m^3$  at a few urban roadside locations. Figure 5.2 shows the annual mean background  $PM_{2.5}$  concentration in 2012.



Figure 5.2: Annual mean background PM<sub>2.5</sub> concentration, 2012 (µg/m<sup>3</sup>).

Index	1	2	3	4	5	6	7	8	9	10	11	12	13
Band	Low	Low	Low	Moderate	Moderate	Moderate	High	High	High	Very High	Hazardous	Very hazardous	Very hazardous
µg/m³ (Defra)	0-88	89-177	178- 266	267-354	355-443	444-532	533- 710	711- 887	888- 1064	1065- 1606	1607-2660	2660-7980	7980+
ppb (Defra)	0-32	33-66	67-99	100-132	133-166	167-199	200- 266	267- 332	333- 399	400- 604	605-1000	1001-3000	3000+

Table 5.1: Sulphur dioxide air quality index values used for analysing air concentration in this work.

The values in bands 1-10 are based on 15-minute averages, but following discussion with PHE and the Department for Health they have been applied to both the hourly and daily averages.

Index	1	2	3	4	5	6	7	8	9	10	11	12	13
Band	Low	Low	Low	Moderate	Moderate	Moderate	High	High	High	Very High	Hazardous	Very hazardous	Very hazardous
µg/m³	0-11	12-23	24-35	36-41	42-47	48-53	54-58	59-64	65-70	71-150	151-250	250-500	500+

Table 5.2: Sulphate aerosol (assumed equivalent to PM2.5) air quality index values used for analysing air concentration in this work.

The values in bands 1-12 are based on 24-hour averages, but following discussion with PHE and the Department for Health they have been applied to the hourly averages also.

# 5.1 Flight level thresholds

Concentrations at flight levels are also presented on the Daily Air Quality Index scales (Tables 5.1 and 5.2). The use of this scale was specified by the Department for Transport, but it should be noted that the levels are relevant to health thresholds at ground level and are not necessarily applicable to understanding any other impacts (e.g. effects on aircraft). Following discussion with DfT and CAA information is only given for concentrations in the Moderate4 band and above.

Data is presented as "hourly" values, which are derived from 1-hour mean air concentrations. 24-hour or "daily" values have not been considered for the flight layers, as this averaging period is far too long compared to the typical time an aircraft might be in the air and encountering a plume. This decision was made following discussion with the Department for Transport. The thresholds for sulphate are based on 24-hour average concentrations so may not be entirely suitable for comparison to hourly averages. All of the statistics and maps are derived from the eighty 6-week scenarios (also referred to as "Periods"), so the hourly values are derived from 80640 (80x6x7x24) 1-hour means

All concentration data at flight levels are given in  $\mu$ g/m<sup>3</sup>. These could be converted to ppb using standard atmospheric pressure and temperature values, but care is needed in interpreting ppb values in the same way when not at the surface.

# 5.2 Points of note regarding sulphate chemistry

In the atmosphere sulphur dioxide (SO<sub>2</sub>) is oxidised by several different chemical reactions. Gas phase oxidation by the hydroxyl radical (OH) is relatively slow, a few percent per hour during daylight. Aqueous phase atmospheric oxidation of sulphur dioxide takes place in cloud and rain droplets which involves rapid oxidation by hydrogen peroxide and ozone, although the ozone pathway is limited by the acidity (pH) of the cloud. The chemical reactions for this process used in NAME and EMEP4UK are summarised in Section 4. All sulphuric acid sulphate is present in particulate form in the atmosphere because it has a very low vapour pressure.

In the atmosphere the major chemical (base) which neutralises the sulphuric acid is ammonia. Ammonia is a gas emitted at the Earth's surface, but is present throughout the troposphere albeit at low concentrations (<1  $\mu$ g/m<sup>3</sup>) in the non-surface atmospheric layers. In the non-surface layer, i.e. in a low ammonia regime, the available ammonia will probably not neutralise all "sulphuric acid" sulphate particles, but the acidity will slowly get neutralised as the "sulphuric acid" sulphate particles scavenge available gas phase ammonia over time.

In the presence of ambient surface ammonia concentrations (typically of the order of  $1-15 \ \mu g/m^3$  at the UK surface) sulphuric acid particles rapidly equilibrate with the ambient ammonia in the atmosphere. The particles exist as an ammonium-sulphate solution or salt. Between the fully neutralised ammonium sulphate and fully acidic sulphuric acid sulphate, there is a range of compositions and acidities, including the salts of ammonium bisulphate and letivocite.

In this project all particulate sulphate is discussed without reference to its neutralisation state, but the reader should note that at the surface and in aged plumes the particles will be much less acidic than freshly formed sulphuric acid particles.

# 6 Analysis of the model outputs of SO<sub>2</sub> and SO<sub>4</sub> for the near-surface

Results from the model simulations conducted with the NAME and EMEP4UK models are presented for the near-surface. The statistics and data presented are those requested by the customer Departments. The report provides an evaluation of the ambient mass concentrations of  $SO_2$  and  $SO_4$  that could be experienced during and following an eruption on Iceland of the type specified, as well as the likelihood of these concentrations.

In the following sections, results from both models for each of the requested outputs are provided. Full sequences of the maps can be found in the related Annexes.

# 6.1 Median, Maximum and Percentiles

The median daily and hourly concentrations derived from the 80 simulations from the two models are within the Low1 Air Quality Index Band for both SO<sub>2</sub> and SO<sub>4</sub> across the whole of the UK and Europe (figures are not presented, as maps are uniformly green). For the 95<sup>th</sup> percentile concentration the UK is within the Low1 band for SO<sub>2</sub> and the Low2 band for SO<sub>4</sub>. The two models show very consistent results for the magnitude and distribution of both species at this percentile (Fig 6.1 and 6.2). Differences in the SO<sub>4</sub> concentrations over the Mediterranean are due to the inclusion of anthropogenic and Etna emissions in EMEP4UK. For SO<sub>2</sub> the 95<sup>th</sup> percentile is so low (<30 µg/m<sup>3</sup>) that a log-scale is needed to reveal the detail in the spatial distribution. The 90<sup>th</sup> percentile outputs provide limited additional information and so are not shown.

The maximum concentration experienced in any one grid square at any time (for both daily and hourly data) in the 80 simulations is depicted in Figures 5 and 6. These maximum values are considerably higher than the 95<sup>th</sup> percentile as would be expected, with both species reaching concentrations in the Very High and above index bands in the outputs from both models. Maximum values in adjacent grid squares are not necessarily from the same simulation, however the apparent plume features displayed in the maximum value maps (Fig 6.3 and 6.4) demonstrate that the maxima, unlike the 95<sup>th</sup> percentile, appear to relate to individual plumes and times. This is true for both models and both species and shows that these maxima are therefore very meteorology and situation specific and that one plume event is generally the cause of the maxima in a particular location. This is further illustrated in the case studies presented in Section 6.7. As such the maxima should be treated as indicative of the concentrations that could be reached more generally in a region. rather than as a specific value for each grid square. These data highlight the sensitivity of the results to the exact meteorological situation and it is worth noting again that the 80 simulations performed here will not have exhaustively explored this variability. They also demonstrate that there is no one period of "worst weather" for affecting the whole of the UK.



Figure 6.1: Maps of 95th percentiles of surface  $SO_2$  concentration for daily and for hourly means on log-scale.



Figure 6.2: Maps of 95th percentiles of surface  $SO_4$  concentration for daily and for hourly means on AQ scale.



Figure 6.3: Map of maximum  $SO_2$  surface concentration for daily and for hourly means on AQ scale.

NAME

EMEP4UK



Figure 6.4: Map of maximum  $SO_4$  surface concentration for daily and for hourly means on AQ scale.

# 6.2 Probability of Threshold Exceedance

The data produced by the models are informative about the probability of air quality thresholds being exceeded, but given the limitations of the study do not provide a full evaluation of the probability. Instead, the results demonstrate the relative frequencies of occurrence – i.e. the number of simulations in which a threshold was exceeded – rather than the probability. These relative frequencies are discussed in this section, but for ease they are also referred to as probabilities.

The probability (relative frequency) of a threshold j being exceeding **at least once** at the surface in a 6-week eruption is calculated for each grid square using the following equation:

$$PA_{j} = \frac{\sum_{i=1}^{80} E_{ij}}{80}$$

Where:

 $PA_j$  = the relative frequency of any time-step in a 6-week eruption period exceeding a threshold j. This is evaluated from the 80 eruption periods considered.  $E_{ij}$  = 0 or 1 depending on whether the threshold j is exceeded at any time-step in period i or not

Maps of the relative frequency with which each threshold is exceeded at the surface for each grid square are given in Annex B for daily and for hourly means. Values are given as a percentage. The lowest value possible in this calculation (other than zero) is 1 in 80 (i.e. in one simulation out of the 80 this threshold was exceeded at least once), which is 1.25%. For all locations, the probability of exceeding the Low1 lower threshold is 100%, as this threshold is set to zero. Table 6.1 lists the highest relative frequency found in any grid square in the UK and Republic of Ireland. The values in Table 6.1 are indicative only and are not necessarily representative of the whole region.

The data show that:

- For daily (24-hour mean) concentrations of SO<sub>2</sub>, most of the UK has a less than 10% probability that the Low2 lower threshold will be exceeded at any time in a 6-week eruption. For parts of Northern Ireland, the percentage is zero, whereas for parts of East Anglia the percentage is up to approximately 14%. Limited parts of the UK have a chance of exceeding the Low3 and Moderate4 thresholds.
- For hourly SO<sub>2</sub> concentrations the probabilities are higher, with all of the UK having up to ~40% chance of exceeding the Low2 threshold. All of the UK has up to a 15% chance of exceeding the Moderate4 threshold. The NAME data suggests that parts of the UK have a small chance of exceeding thresholds up to and including VeryHigh10 (see maps for spatial detail).
- SO<sub>4</sub> shows higher frequencies of exceeding AQI thresholds than SO<sub>2</sub>. The SO<sub>4</sub> data also show a north-south gradient, which is not apparent in the SO<sub>2</sub> data.
- For daily (24-hour mean) concentrations of SO<sub>4</sub>, parts of southern England and Wales exceed the Low2 lower threshold at some time in 90-100% of simulations. Values in Scotland and part of Northern Ireland are lower for this threshold, but are still between 60-70%. For the Moderate4 threshold numbers reduce to 30-40% for most of the UK (20-30% in Scotland). As the

thresholds increase, relative frequencies decrease, but even at Very High10 the majority of the UK exceeds this level on a single day in an eruption period in 10-20% of the simulations.

For hourly SO<sub>4</sub> concentrations, probabilities are substantially greater than the daily equivalents. All of England, Wales and Northern Ireland and most of Scotland have probabilities of 80-100% of exceeding the Moderate4 threshold and 60-80% of exceeding the High7 threshold. Even at the VeryHigh 10 level, large areas of the south of the UK exceed this concentration in >60% of simulations. Of note is that the whole of the UK has a (non-zero) probability of exceeding the Very Hazardous levels in any 6-week eruption as simulated here.

AQI	Maximu	Maximum frequency of exceedance anywhere in UK and Rep. Ireland						
Level	Daily SC	<b>)</b> <sub>2</sub>	Hourly S	SO <sub>2</sub>	Daily SC	<b>D</b> 4	Hourly S	SO <sub>4</sub>
	NAME	EMEP	NAME	EMEP	NAME	EMEP	NAME	EMEP
		4UK		4UK		4UK		4UK
1	100	100	100	100	100	100	100	100
2	13.75	8.75	43.75	25.0	87.5	81.25	100	98.75
3	3.75	2.5	21.25	10.0	61.25	46.25	100	80.0
4	2.5	1.25	16.25	7.5	43.75	26.25	95.0	55.0
5	1.25	0	11.25	5.0	37.5	18.75	91.25	40.0
6	1.25	0	7.5	2.5	33.75	16.25	88.75	33.75
7	0	0	5.0	1.25	31.75	15.0	83.75	28.75
8	0	0	3.75	0	27.5	11.25	78.75	25.0
9	0	0	3.75	0	26.25	8.75	75.0	21.25
10	0	0	2.5	0	23.75	7.5	71.25	20.0
11	0	0	0	0	12.5	1.25	33.75	2.5
12	0	0	0	0	8.75	0	22.5	0
13	0	0	0	0	3.75	0	11.25	0

Table 6.1: Maximum percentage relative frequencies of exceeding the lower threshold of each of the AQI bands at any time in a 6-week eruption period for a grid square anywhere in the UK and Ireland region. Note: the Low1 thresholds are zero, so the associated values (in italics) are not necessarily very meaningful.

#### 6.3 Consecutive Duration of Exceedances

The consecutive length of time that each grid square remains above each of the Air Quality thresholds has been calculated for each of the 80 simulations. The maximum consecutive duration in any of the simulations has then been extracted for each grid square. This gives an indication of the "worst-case" duration of exposure that could be expected during 'volcanic air pollution episodes'. Note that one 6-week eruption period could result in multiple air pollution episodes. For context, the total time modelled was 80640 hours and 3360 days, of which 67200 and 2800 respectively were during emission periods.

Maps of the maximum number of consecutive days/hours of exceedance of a certain threshold at the surface are given in Annex B. Table 6.2 lists the highest consecutive duration found in any grid square in the UK and Republic of Ireland for both averaging periods and species. The values in Table 6.2 are indicative only and are not necessarily representative of the whole region. Note: these values are the time spent at and above the AQI level lower threshold, not the time spent within each AQI band. These data reveal that:

- Maximum exceedance durations for SO<sub>2</sub> are much shorter than for SO<sub>4</sub>.
- For SO<sub>2</sub> there are no periods when the lower Low 3 threshold (178 μg/m<sup>3</sup>) is exceeded for two consecutive days based on modelled 24-hour means.
- For the hourly means for SO<sub>2</sub>, the majority of the UK could experience a maximum 2-5 consecutive hours above the Moderate4 threshold. Parts of the UK, including East Anglia, the east coast of England and also the Scottish Islands, could experience similar maximum durations above the High7 thresholds.
- There are no incidences with consecutive hours at the Hazardous11 level or above for SO<sub>2</sub>.
- For SO<sub>4</sub>, all of the UK could experience 2+ consecutive days (based on 24-hour means) above the High7 threshold of 54 μg/m<sup>3</sup>. Certain areas of the UK also experience 2+ days above the Hazardous11 threshold of 151 μg/m<sup>3</sup>. Parts of the UK could also experience up to 13 consecutive days where at least the Low1 threshold is continuously exceeded.
- The hourly mean data for SO<sub>4</sub> shows a similar pattern to the daily means, but here more or less the whole of the UK experiences two or more consecutive hours above the Very Hazardous threshold of 500 μg/m<sup>3</sup>.
- One of the complications with looking at the maximum length of consecutive time periods is that the results can be strongly biased by any outlier or extreme cases. For example, the hourly SO<sub>4</sub> values to the south west of the UK in the NAME data appear to be dominated by a small number of cases with the plume to the west of Spain. Whilst these data are still useful for understanding the extremes, they do not necessarily reflect the situation across all of the scenarios. The values in Table 6.2 for SO<sub>4</sub> should be considered a worst case as the actual modelled values for hourly SO<sub>4</sub> are much smaller over most of the UK.

AQI level	Daily SO <sub>2</sub>	Hourly SO <sub>2</sub>	Daily SO <sub>4</sub>	Hourly SO <sub>4</sub>
1	42	1008	42	1008
2	3	41	12	199
3	<2	31	11	148
4		26	9	120
5		23	9	117
6		18	8	116
7		15	7	114
8		14	7	114
9		10	6	114
10		7	6	114
11		0	5	75
12		0	4	58
13		0	<2	52

Table 6.2: Maximum consecutive duration above each threshold at any time in a 6-week eruption period for a grid square anywhere in the UK and Ireland region in the NAME model results. Note: the Low1 thresholds are zero, so the associated values (in italics) are not necessarily very meaningful.

#### 6.4 Mean Total Time Thresholds are Exceeded

The mean numbers of hours and days (based on 24-hour mean concentration) a certain threshold is exceeded at the surface in a 6-week eruption period have been derived from the total number of exceeded times in the whole 80 simulation periods. The following equation has been applied to each modelled grid square:

$$\overline{T}_j = \frac{\sum_{i=1}^{80} T_{ij}}{80}$$

Where:

 $\overline{T}_{j}$  = the mean number of time-steps above a threshold j per 6-week eruption period

 $T_{ij}$  = the total number of time-steps above threshold j in period i

Maps of the calculated mean number of hours and days are given in Annex B for each threshold. These give an indication of the total time a threshold could be exceeded during a 6-week eruption (note that these are not consecutive times). For the hourly data, only mean values above 1 hour are plotted, for the daily values, smaller fractions (less than 1 day) are allowed. Table 6.3 lists the highest mean total-time seen in any of the grid boxes over the UK and Ireland. These data reveal that:

- For SO<sub>2</sub>, the Low2 and Low3 thresholds are only exceeded for a very short number of days and hours on average in both models. The 89 µg/m<sup>3</sup> (Low2) threshold is exceeded on less than one day per eruption period (for both hourly and 24-hour mean concentrations). In NAME the 178 µg/m<sup>3</sup> (Low3) threshold is only exceeded in the east of the UK for less than 2 hours in any period in the hourly data. In EMEP4UK this Low3 threshold is not exceeded anywhere over the UK for more than hour.
- The spatial distribution over the UK in both models suggests that the east of England is more likely to experience the longest times of SO<sub>2</sub> above these thresholds.
- Analysis of the distribution for the lower WHO limit of 20 µg/m<sup>3</sup> (Fig 6.5), which shows the finer level geographical detail on the European scale, reveals that the mean total number of hours that this threshold is exceeded increases with distance away from Iceland and is higher over much of continental Europe compared to the UK. The contribution from anthropogenic emissions of sulphur dioxide is clearly evident at these low concentrations in the EMEP4UK data. (The reason for the elevated area on the east coast of Spain in the NAME results is unclear and needs further investigation.)



Mean total number of hours per period above 20.0 ug/m3



- For SO<sub>4</sub>, most of the UK experiences time above all of the thresholds up to and including the Very Hazardous 12 threshold of 250 µg/m<sup>3</sup> in the NAME data although this is on the order of only a few hours in total at this level. In the EMEP4UK data no part of the UK experiences any time (greater than an hour) at or above the Very Hazardous 11 threshold of 151 µg/m<sup>3</sup>. Notably, both models suggest that all of England and Wales will experience some time over the Very High 10 threshold.
- Over the UK there is a clear north-south gradient in the total time spent above each threshold shown by both models. Southern and eastern regions experience longer times than those in the North. This is in part due to the conversion time for SO<sub>2</sub> to SO<sub>4</sub>, which means concentrations of SO<sub>4</sub> increase with distance away from Iceland.
- On the European scale, the spatial distribution of the mean number of hours of SO<sub>4</sub> at the lowest end of the AQI concentration scale is different to that of SO<sub>2</sub>. The highest SO<sub>4</sub> totals occur to the west and north of Spain over the Atlantic and to the East of Spain over the Mediterranean (Fig 6.6), whereas the highest SO<sub>2</sub> totals are more over the European continent (Fig 6.5).



Mean total number of hours per period above 12.0 ug/m3

Figure 6.6: Mean total number of hours (out of 1008 hours per 6-week eruption period) above the Low2 12  $\mu$ g/m<sup>3</sup> SO<sub>4</sub> limit as simulated using NAME (top panel) and EMEP4UK (bottom panel), showing the finer distribution detail.

AQI	Hourly S	SO <sub>2</sub>	Hourly S	SO <sub>4</sub>
index	NAME	EMEP 4UK	NAME	EMEP 4UK
1	1008	1008	1008	1008
2	4.3	2.6	75.5	129
3	1.4	0.8	39.8	34
4	<1	<1	25.7	16
5	<1	<1	21.7	12
6	<1	<1	18.3	9
7	<1	<1	15.6	8
8	<1	<1	13.8	6
9	<1	<1	12.1	5
10	<1	<1	10.7	4
11	-	-	4.0	<1
12	-	-	2.6	<1
13	-	-	1.5	-

Table 6.3: The highest number of mean total hours (hours >1) above the lower threshold of each of the AQI bands simulated in a grid square anywhere over the UK and Ireland. Note: the Low1 thresholds are zero, so the associated values (in italics) are not necessarily very meaningful.

# 6.5 Arrival Times

Seven regional boxes have been constructed to represent the UK and Republic of Ireland (Fig 6.7(a)). These have been used to determine the fastest arrival of a plume after the start of the eruption in each of the 80 scenarios. The first appearance of the plume is taken to be when the Low2 AQI band lower threshold is exceeded, which for  $SO_2$  is 89 µg/m<sup>3</sup>. Note that the arrival times of plumes from different eruptive episodes during the eruption scenario cannot be easily calculated and could be faster.

Using the NAME model data the quickest time (in hours) of arrival of a plume to each of the boxes has been extracted and the mean time of arrival has also been calculated from all of the 80 scenarios modelled (Table 6.4). Each box has been considered independently. The results show that a detectable (>89  $\mu$ g/m<sup>3</sup> SO<sub>2</sub>) plume could reach the northern extremities of the UK in just over one day (29 hours), but that parts of the country more distant from Iceland might have slightly longer warning times, before the plume arrives, of around 2.5+ days. The case studies provide further detail on particular episodes.

The mean time of arrival calculated over all of the simulation periods is also given in Table 6.4; this gives a much longer time of 18-23 days. However analysis of the spread of first arrival times across all of the 80 simulations (Fig 6.7(b)) shows that this mean value is not particularly representative or meaningful.

Although this analysis is currently only carried out for results from the NAME model we do not expect substantially different findings from the EMEP4UK model, because the time of arrival is mainly driven by meteorology (3D wind speed and direction), which in the case of the EMEP4UK model is derived from re-analysis NWP data.

Вох	Location	Quickest time (hours)	Mean time (hours)
1	Northern Ireland	29	522
2	Republic of Ireland	67	502
3	Scotland	29	437
4	Wales	64	571
5	Northern England	75	490
6	Central & Eastern England	64	518
7	Southern England	66	494

Table 6.4: Arrival time results from the NAME model. The mean time is calculated from onlythose periods when the plume reached the box.



Figure 6.7: (a) Map of the seven boxes used to determine the first arrival-time of the plume over the UK. (b) Frequency distribution of the first arrival-time in all of the 80 simulation periods for each box. The shaded area denotes the range of mean values in Table 6.4.

## 6.6 Population Weighted Mean Concentration

Population weighted mean concentrations for the UK have been calculated at the request of Defra for each 6-week simulation period using the following methodology:

- Population data for 2011 provided by Defra (via Riccardo-AEA) in the form of the number of people per grid square have been re-gridded from their original 1 km x 1 km resolution to 25 km x 25 km resolution to compare to the NAME model output (Fig 6.8).
- For each grid square (n), the modelled 6-week period mean concentration  $(\overline{C_n})$  has been multiplied by the population in the grid square  $(p_n)$ . This gives one value per grid square.
- These individual grid square values have been summed to get a total over all the UK.
- This total is then divided by the total UK population ( $P_{1} \sim 63$  million) to give one number, which is the population weighted mean concentration, for that 6week period.

This can be represented by the following equation:



Figure 6.8: (a) The original 1 km resolution UK population data and (b) the regridded 25 km resolution data. Note the different contour scales used. Both scales show the number of people per grid square.

regridded population data (count) 1E+04 1E+05

Dara Mm - 06+00, Max - 46+01

1E+06

1E+03

1E+ 02

This has been repeated for all 80 simulation periods. The resulting maximum and mean population weighted mean concentrations (over all 80 periods) are given in Table 6.5 and the distributions of the means calculated from each simulation period are shown in Figure 6.9. Because the durations of plume exposure are relatively short compared to the 6-week period over which the mean is calculated, the 6-week period mean concentrations are relatively low and have a strong influence on these calculations.

Species	Max pop weighted mean (µg/m³)	Mean pop weighted mean (µg/m³)	
SO <sub>2</sub>	10.9	1.8	
SO <sub>4</sub>	12.5	3.5	



Table 6.5: Population weighted mean values for SO<sub>2</sub> and SO<sub>4</sub> derived from the 80 simulations.



Figure 6.9: Distribution of the population weighted mean concentrations calculated for each of the 80 simulation periods for (a) SO<sub>2</sub>, (b) SO<sub>4</sub>.

#### 6.7 Case-studies of high impact periods over the UK

A huge amount of data has been produced in this work and the summary results presented so far provide no detail on specific periods or the evolution of pollution episodes. Analysis of the raw hourly outputs from the models shows that the plumes are highly transient in nature and their location is strongly controlled by the meteorology. To provide more detail on the time evolution of the SO<sub>2</sub> and SO<sub>4</sub> plumes over the UK, a selection of case studies are presented. These have been selected based on analysis of the NAME model data.

Potential case-study episodes were chosen by looking at the daily maximum concentrations of each species for the near-surface. A short-list was compiled of all times when these values were significantly elevated. For each episode on the short list the hourly sequences were then examined in more detail in order to determine a limited number of case-studies that represented significant episodes in terms of both magnitude of the concentration, extent and duration of the episode. In addition, the highest population weighted mean periods were examined, which for SO<sub>2</sub> are in periods 6, 20 and 26 and for SO<sub>4</sub> are in periods 7, 53 and 68. These are coincident with the periods when the most significant plume concentrations were experienced over the UK.

The selected episodes were compared to the maxima data from the full sequence to ensure that they corresponded to detail on these maps. This resulted in Period 6 being chosen as one of the  $SO_2$  case-studies along with a further case-study for  $SO_2$  (Period 26) and one for  $SO_4$  (Period 53).

To enable the uncertainty analysis conducted in Section 8.2, three sites have been identified for extraction of time series data from the model output. The locations of these sites are shown in Figure 6.10 and they are referred to as North, East and West in the following text. Major population centres have not been chosen to prevent over-interpretation of the data at specific locations.



Figure 6.10: The three sites used for analysis.

#### 6.7.1 Case-Study 1: Elevated near-surface SO<sub>2</sub>

This case-study occurs in Period 6, which begins on 30/07/2003. The UK episode starts on Day 18 of this eruption period, which is the 16/08/2003, and lasts for about 2 days. Figure 6.11 shows the NAME simulated maximum hourly value of SO<sub>2</sub> simulated in this period for each grid square. A large swath of southern England and Wales is affected by concentrations in the moderate and high index levels in this Period. Moderate levels are also experienced in Northern Ireland. Maximum concentrations of SO<sub>4</sub> experienced are also elevated (within the Very High10 and Hazardous11 AQI bands) during this period, but the values are not extreme compared to other periods that are used as case studies.



Figure 6.11: Maximum hourly concentration over simulation Period 6

More detail on the temporal evolution of the  $SO_2$  cloud is given in Figure 6.12. In the NAME data (Fig 6.12(a)) the plume appears off the east coast of England at the start of the episode and over the following 24 hours is transported to the west passing over London, the home counties, south-west England and south Wales, before being dispersed north-westwards over the Irish Sea to Ireland and Northern Ireland.

The maximum NAME simulated daily concentrations for a large zone across the south of the UK were within the Moderate4 band (Fig 6.13) and occur for one day only. The maximum NAME hourly SO<sub>2</sub> concentrations for a large area of the southern UK are within the High index bands (Fig 6.12(a)) with the maximum simulated anywhere over the UK in this period being 909  $\mu$ g/m<sup>3</sup>, which is in the High9 index band.

The maximum values simulated by NAME at the three analysis sites are: North 224  $\mu$ g/m<sup>3</sup> (15:00 16 Aug), East 306  $\mu$ g/m<sup>3</sup> (11:00 16 Aug), and West 325  $\mu$ g/m<sup>3</sup> (21:00 16 Aug). Figure 6.14(a) shows the time-series of SO<sub>2</sub> air concentrations simulated at each of the three sites during the episode. The distribution of these hourly concentrations with respect to the air quality index bands (Fig 6.14(b)) demonstrates that the majority of the time is spent within the three Low index bands for these sites.

The timing of the plume arrival in the EMEP4UK data (Fig 6.12(b)) is very similar to the NAME model, however the EMEP4UK SO<sub>2</sub> surface concentrations are lower than those modelled by NAME. For example at 18:00 on 16 August the maximum surface

concentration from the EMEP4UK model is ~200  $\mu$ g/m<sup>3</sup> whereas the NAME model is ~ 550  $\mu$ g/m<sup>3</sup>. Further detailed analyses are required to properly explain these differences.

August 2003 was very warm and sunny across all parts of the UK and rainfall was well below or exceptionally below average across the majority of the UK, which would have reduced wash-out of any pollution from the atmosphere. From the  $3^{rd}$  to the  $13^{th}$  August a record-breaking heat wave occurred especially in the Midlands, central southern and south-east England and East Anglia as high pressure took control bringing prolonged sunshine to most areas and breaking UK temperature records. High pressure remained dominant over the south-east during the identified elevated SO<sub>2</sub> period (Fig 6.15), meaning that the gas cloud would have remained near the surface and been poorly dispersed. Light easterly winds near the surface led to the movement of the plume across the south of England to Wales and then into the Republic of Ireland.

Air history maps have been produced using NAME for the three analysis sites for the hours when their simulated concentrations were maximum. These maps show where the air reaching the near-surface at each site originated from. Due to the travel time and complexity of the air masses, simple deterministic backwards trajectories are not sufficient to understand the origins of the SO<sub>2</sub> and full stochastic dispersion simulations are needed.

The air history maps (Fig 6.16) reveal that the SO<sub>2</sub> plume that reaches the three sites in this episode is primarily related to emissions from Iceland on the 12 Aug (and also the 13th for the East site), which was Period 6 day 13. On this day the SO<sub>2</sub> emissions were 2.22 Tg, which happens to be the second largest day of emissions in the emissions sequence and also the day with the third largest emissions into the troposphere (Table 6.6). By producing air history maps for different originating vertical levels we can also determine that in this instance it was primarily emissions from below 6000 m asl over the vent that contributed to the plume over the UK during this episode. The travel time for the plume to the UK in this episode is 3-4 days.

Conditions associated with heat waves often lead to heightened air pollution levels originating from routine emissions. Notably August 2003 was also a period of poor air quality across the UK, with elevated ozone and PM10 contributing to excess mortality in addition to that caused by the high temperatures. It is interesting to note that in this case study, the simulated volcanic release and consequent elevated SO<sub>2</sub> and SO<sub>4</sub> would be just after the significant anthropogenic pollution episode that occurred on 5-14 August, potentially prolonging the high risk period from a health perspective. The routine anthropogenic emissions have been included in the EMEP4UK simulations so the levels of other species could be evaluated, making this is an interesting episode for further study.

Day in eruption	Tropospheric emission
sequence	(Tg)
23	1.778
7	1.640
13	1.621
1	1.533
8	1.482

Table 6.6: Top five days in the eruption sequence for SO<sub>2</sub> emissions into the troposphere.



Figure 6.12(a): NAME hourly mean  $SO_2$  air concentrations for every six hours during the episode, plotted using the AQ index scale.



Figure 6.12(b): EMEP4UK hourly mean  $SO_2$  air concentrations for every six hours during the episode.



Figure 6.13(a): NAME daily  $SO_2$  means for days 17-20 in Period6 plotted using the AQ index scale.



Figure 6.13(b): EMEP4UK Daily SO<sub>2</sub> means for days 17-20 in Period6 plotted using the AQ index scale.



Figure 6.14: (a) Hourly time series of modelled SO<sub>2</sub> air concentrations at the three analysis sites, (b) frequency distribution of the hourly concentrations by AQ index band for the duration of the episode (only hours with non-zero concentrations are included).



Figure 6.15: Weather chart for 12:00 UTC on 15 August 2003, showing the high pressure system that had been controlling the UK's weather up to this point.



Figure 6.16: Snapshots every 24 hours showing the previous locations of the air mass that reached the East site between 10:00 and 11:00 on 16 Aug 2003. These maps are a vertically integrated SO<sub>2</sub> mass (an atmospheric "total column"), so provide no information on height distribution or air concentration. Maps are arranged from left to right going back in time.

#### 6.7.2 Case-Study 2: "Very High" near-surface SO<sub>2</sub>

This case-study occurs in Period 26, which begins on 07/03/2006. During this Period the UK experiences two episodes of elevated SO<sub>2</sub>, separated by around 12 days. The first episode lasts about two days and is mainly characterised by hourly concentrations in the Low bands, although moderate levels are briefly reached in parts of Scotland. In the second episode (which lasts around 36 hours) the maximum concentration simulated by NAME in the UK region is 1566  $\mu$ g/m<sup>3</sup>. This is within the Very High 10 band and is significantly higher than that seen in Case-Study 1. The NAME modelled hourly maximum concentrations (Fig 6.17) show that the bulk of the High concentration SO<sub>2</sub> plume is over Central and Eastern England. Figure 6.17 also shows that the Republic of Ireland is strongly affected in this episode. The temporal evolution of the second episode is depicted in Figure 6.18.



Figure 6.17: Maximum hourly  $SO_2$  concentration over simulation Period 26.

Similar to the Case Study 1 episode, the timing of the plume is comparable between the NAME (Fig 6.18(a)) and EMEP4UK (Fig 6.18(b)) models, showing a plume to the west of and over Ireland at 00:00 and 06:00 on 5 April and a plume over the east of England at 12:00 and 18:00. However, EMEP4UK SO<sub>2</sub> surface concentrations are lower than NAME. For example at 18:00 on the 5 April the maximum surface concentration from the EMEP4UK model is ~200  $\mu$ g/m<sup>3</sup> whereas for the NAME model it is ~800  $\mu$ g/m<sup>3</sup>. Further analyses are required to properly explain the differences.

During the first episode  $SO_2$  concentrations at the three analysis sites remain in the Low bands, but during the second episode concentrations at the East site reach 1070  $\mu$ g/m<sup>3</sup> in the NAME simulation, which is in the Very High10 index band (Fig 6.19). Peak NAME concentrations at the other two sites are 396  $\mu$ g/m<sup>3</sup> at the West site and 279  $\mu$ g/m<sup>3</sup> at the North site. The times of the arrival of the plume and the peak concentrations are predominantly in the Low1 category and reach the Moderate band over East Anglia during only one day (Fig 6.20), which is day 30 in the 6-week simulation Period. Although a plume is present over the West site on the 04/04/2006, its presence is short-lived and daily means are within Low1 everywhere across the UK on this day. The frequency distribution of the hourly values (Fig 6.19(b)) demonstrates that the majority of hourly values are within the Low bands.

NAME modelled air history maps suggest that the SO<sub>2</sub> plume simulated over the east of England on the 5 April is due to emissions from Iceland on the 3 April (Fig 6.21). This represents a much faster travel time than in case-study 1. The 3 April is Day 27 in the emission sequence for this Period and is a day with only 3 lower level emission layers, all of which are in the troposphere. Total emissions on this day are relatively low compared to other days at 0.51 Tg, but all are emitted below 4.5 km asl. Transport to the UK also occurs predominantly below this altitude and wind information for this altitude range is given in Table 6.7. Compared to other emissions, this amount is an order of magnitude higher than the SO<sub>2</sub> emitted in this height range on the more vigorous eruptions days. Of the days with only low-level emissions (below 5 km), day 27 is the fourth highest emission (Table 6.8). Further consideration of the impact of the emissions during this period is given in Section 8.2.

Transport of the plume from Iceland is linked to the south-eastwards passage of a low pressure centre just to the south of Iceland on the 3<sup>rd</sup> and 4<sup>th</sup>. This feature can be clearly identified in the air history maps in Figure 6.21. On the 5<sup>th</sup> two cold fronts ahead of the low cleared southwards over the UK (Fig 6.22). In reality this introduced much colder conditions to the country, but in the simulation they additionally have the effect of transporting the plume southwards and bringing it towards the surface.



Figure 6.18(a): NAME hourly mean  $SO_2$  air concentrations for every six hours during the episode, plotted using the AQ index scale.



Figure 6.18(b): EMEP4UK hourly mean  $SO_2$  air concentrations for every six hours during the episode.



Figure 6.19: (a) Hourly time series of modelled SO<sub>2</sub> air concentrations at the three analysis sites, (b) frequency distribution of the hourly concentrations by AQ index band for the duration of the episode (only hours with non-zero concentrations are included).



Figure 6.20(a): NAME daily SO<sub>2</sub> means for Period 26 plotted using the AQ index scale.



Figure 6.220(b): EMEP4UK daily SO<sub>2</sub> means for Period 26 plotted using the AQ index scale.

Height (m asl)	Mean Wind speed (m/s)	Max wind speed (m/s)	Mean wind direction (degrees)
1000	8.7	15.5	265
2000	10.2	14.6	300
3000	11.7	16.6	306
4000	13.4	16.7	311
5000	16.0	19.9	320

Table 6.7: Wind information derived from the Met Office's UM Global model for the period 12 UTC on 3 April 2008 to 12UTC on 4 April 2008 for the location N62, W12.

Day in eruption sequence	Emissions below 5 km asl (Tg)
17	0.570
26	0.565
16	0.557
27	0.509
15	0.480

Table 6.8: Top five days with the highest total emissions below 5 km. These are all days represented by 3-layers in the emission source.



Figure 6.21: The history of the air mass that reached the East site at its maximum concentration time. These maps are a vertically integrated SO<sub>2</sub> mass (an atmospheric "total column"), so provide no information on height distribution or air concentration. Maps are arranged from left to right going back in time.



Figure 6.22: Analysis chart for 00:00 UTC on 5 April 2008 showing the northerly air flow and cold fronts moving over the UK at this time. The Low pressure marked as 1001 behind the northern-most front is the remains of the low pressure centre that passed close to Iceland on the preceding days.
#### 6.7.3 Case-Study 3: Elevated near-surface SO<sub>4</sub>

This case-study occurs in Period 53, which starts on 03/07/2009. The episode starts on around day 30 in this eruption period, which is the 1/8/2009, and lasts for about 36 hours. Figure 6.23(a) shows the maximum hourly value of SO<sub>4</sub> simulated by NAME in this period for each grid square. Very Hazardous (black) levels are reached over much of central, east and south-east England during this episode, but elevated levels affect almost the entire country at some time. Figure 6.23(b) demonstrates that minor increases in SO<sub>2</sub> are also simulated by NAME in similar areas during this period, but that Moderate4 concentration is only reached in a small area of the Midlands.



Figure 6.23: Maximum hourly concentrations of (a) SO<sub>4</sub> and (b) SO<sub>2</sub> during simulation Period 53 from NAME.

More detail of the temporal evolution of the  $SO_4$  cloud is given in Figures 6.24 and 6.25. In this case study, the plume arrives off the west coast of Ireland and is then transported to the east across Ireland to affect most of the UK within the following 24 hours. This transport corresponds to a period between the passage of two frontal systems across the UK from west to east.

The maximum hourly SO<sub>4</sub> concentration over the UK domain simulated by NAME in this period is 1255  $\mu$ g/m<sup>3</sup>, which is in the Very Hazardous index 13 level. The maximum NAME values simulated at the three sites are: North 343  $\mu$ g/m<sup>3</sup>, East 869  $\mu$ g/m<sup>3</sup>, West 323  $\mu$ g/m<sup>3</sup>. The frequency distribution of the hourly values at the three sites (Fig 6.26) demonstrates that although the majority (~50%) of hourly values are within the Low bands, there is a significant proportion in the Very High, Hazardous and Very Hazardous bands, particularly for the East site (~40%), during this episode.

The SO<sub>4</sub> surface concentrations calculated by the EMEP4UK model are generally lower than those reported by NAME. In addition, in contrast to the SO<sub>2</sub> case studies where the spatial distribution of the hourly plume was comparable between the two models, there are more noticeable differences between the modelled distributions. In particular EMEP4UK does not simulate SO<sub>4</sub> levels above Low over Ireland or in the north west of the UK at the start of the episode. However, both models simulate a plume over the East of England and the North Sea in the later phase, suggesting that the meteorological influence is similar in both models.

NAME modelled air history maps suggest that the  $SO_4$  plume simulated over the east of England on the 3 August is related to emissions of  $SO_2$  from Iceland on the  $30^{th}$ 

July, with contributions from emissions of the  $28^{th}$  and  $29^{th}$  (Fig 6.27). This allows 4-7 days over which chemical conversion of SO<sub>2</sub> to SO<sub>4</sub> can occur in this particular plume.



Figure 6.24(a): NAME daily mean  $SO_4$  air concentrations for 6 days during the episode.



Figure 6.24(b): EMEP4UK daily mean SO<sub>4</sub> air concentrations for 6 days during the episode.



Figure 6.25(a): NAME hourly mean  $SO_4$  air concentrations for every 6 hours during the episode.



Figure 6.25(b): EMEP4UK hourly mean SO<sub>4</sub> air concentrations for every 6 hours during the episode.



Figure 6.26: (a) Hourly time series of modelled SO<sub>4</sub> air concentrations at the three analysis sites, (b) frequency distribution of the hourly concentrations by AQ index band for the duration of the episode (only hours with non-zero concentrations are included).



Figure 6.27: The history of the air mass that reached the East site at its maximum SO<sub>4</sub> concentration time in Period 53. These maps are a vertically integrated mass of a tracer species (an atmospheric "total column"), so provide no information on height distribution or air concentration. Maps are arranged from left to right going back in time.

#### 6.8 European summary

The NAME and EMEP4UK simulations conducted for this work covered a domain that encompassed Europe, some of North Africa and a large part of the North Atlantic. Within the constraints of this project there is not the capacity to present or analyse the data for this large domain. However it is pertinent to touch on the broader European data because:

- 1. In terms of the modelling study, impacts seen anywhere in Northern Europe would also be possible in the any other part of Northern Europe including the UK.
- 2. Wider European impacts will have a bearing on UK interests.

In order to evaluate the uncertainty in the transport, deposition and therefore impact of an effusive Icelandic eruption, this work has used two models and considered 80 meteorological evolutions spread-out over 10 years. The number of simulations was driven by what was practical within the scope of this work, it does however underrepresent the potential number of emission and meteorological evolutions and therefore may misrepresent the level of species that could be encountered during such an event. The scale of the event and the underlying atmospheric transport mean that concentrations seen in most of Northern Europe could be seen in any other part of Northern Europe. Therefore by considering the wider domain it is possible to gain a better idea of the potential concentrations.

Though limited in scope, here we consider 7 sites across Europe. These consist of the 3 sites within the UK, as used in the case studies, and 4 additional sites one each in Spain, France, Germany and Norway (Fig 6.28). Maximum concentrations at these sites (Table 6.9) show a wider range of concentrations across Europe than just the UK. For the sites chosen the highest SO<sub>2</sub> value is seen at the UK East site while all other sites show considerably lower values. For SO<sub>4</sub> the Norway (closer to Iceland) site has the highest value, being 40% higher than any other, and Germany (further from Iceland) shows values comparable to the highest UK site. In terms of the frequency of exceedances of the air quality bands, all sites, while showing a range, exhibit a similar distribution for both  $SO_2$  and  $SO_4$  (Fig 6.29). Figure 6.29 also demonstrates that the use of the AQI banding can give misleading results. The apparent peak for SO<sub>4</sub> in index band 10 in Figure 6.29(b) is due to the non-linear concentration scale used to define the index bands, which is particularly pronounced at and above the Very High10 band (refer to Table 5.2 for details). If these frequencies of highest concentrations are plotted on a linear scale (Fig 6.29(c)) a much more intuitive distribution is seen, with a peak at the "lower" concentrations and a tail in the "higher" concentrations.

Overall these data show, in a limited way, that considerable variability is possible across Northern Europe. This, in part, illustrates the uncertainty that is also present within the results for any specific country.



Figure 6.28: Map of UK and wider European sites used in analysis

Site	Max SO <sub>2</sub>	Max SO <sub>4</sub>
	concentration	concentration
UK North	448.6	1339.7
UK East	1069.8	868.8
UK West	396.0	1242.3
Spain	462.9	470.8
France	306.7	685.2
Germany	267.4	1283.4
Norway	483.6	1895.8

Table 6.9: Maximum hourly  $SO_2$  and  $SO_4$  values per analysis site over the whole 80 simulations.

Figure 6.29 (*next page*): Frequency distribution of the maximum concentrations in each period at each site (i.e. from a sequence of 80 maximums). Plotted by AQ band for (a)  $SO_2$  and (b)  $SO_4$ . (c) Plotted by linear spacing for  $SO_4$ .



# 7 Analysis of the model outputs of SO<sub>2</sub> and SO<sub>4</sub> for aviation purposes

Results from the model simulations conducted with the NAME and EMEP4UK models are presented for a limited range of flight altitudes. The statistics and data presented are those requested by the customer Departments. The report provides an evaluation of the ambient mass concentrations of  $SO_2$  and  $SO_4$  that could be experienced during and following an eruption on Iceland of the type specified, as well as the likelihood of these concentrations. Results are presented for the flight levels pertinent to aviation listed in Table 4.2, which were specified by the Department for Transport.

In the following sections, results from both models for each of the requested outputs are provided. Full sequences of the maps can be found in the related Annexes.

#### 7.1 Median, Maximum and Percentiles

The median hourly concentrations derived from the 80 simulations from the two models for both  $SO_2$  and  $SO_4$  are within the Low1 Air Quality Index Band across the whole of the UK and Europe (figures are not presented, as maps are uniformly green).

For the 95<sup>th</sup> percentile concentration, the UK and continental Europe are within the Low1 band for SO<sub>2</sub> and the Low2 band for SO<sub>4</sub> at the lower flight altitudes (Fig 7.1 and Fig 7.2). This is consistent with the results for the near-surface. The two models show consistent results for the magnitude and distribution of both species near the UK at these lower flight levels, but the influence of European anthropogenic emissions and emissions from Etna can be seen in the EMEP4UK results.

At the upper flight level (FL370), ambient  $SO_2$  concentrations at the 95<sup>th</sup> percentile are very similar between the two models, with highest concentrations near Iceland which is due to the volcanic emissions at this height. The dispersion of the plume to the east of Iceland in Figure 1 reflects the dominant westerly air flow at these altitudes. The 95<sup>th</sup> percentile concentrations of  $SO_4$  (Fig 7.2) remain below 12 µg/m<sup>3</sup> (the Low2 lower threshold) at this upper altitude. This is due to the variability in transport of the plume on the conversion timescales of  $SO_2$  to  $SO_4$ .

The maximum concentration experienced in any one grid square at any hour in the 80 simulations is shown in Figures 7.3 and 7.4 for the six flight levels. These maximum concentrations are considerably higher than the 95<sup>th</sup> percentile concentrations, as would be expected, with both species reaching concentrations in the Very High index bands and above in the outputs from both models. Maximum concentrations in adjacent grid squares are not necessarily from the same simulation, although the apparent plume features displayed in all the maps demonstrate that the maxima, unlike the 95<sup>th</sup> percentile, appear to relate to individual plumes and simulations. This is true for both models, both species and all flight levels, and shows that these maxima are therefore very meteorology and situation specific. The conclusion is that one plume event is generally the cause of the maxima in a particular area or small region. As such the maxima should be treated as indicative of the concentrations that could be reached more generally in a region, rather than as a specific value for each grid square. This is confirmed by the fact that although the specific spatial distributions are somewhat different between the two models at the FL010 and FL370 levels, the maximum concentrations of SO<sub>2</sub> are very similar (Fig. 7.3).

These data highlight the sensitivity of the results to the exact meteorological situation and it is worth noting again that the 80 simulations performed here will not have exhaustively explored this variability. They also demonstrate that there is no one period of "worst weather" for affecting the whole of the UK.

The results highlight that it may be worthwhile for an impact assessment to also analyse and use concentrations at the 99<sup>th</sup> percentile for example.



Figure 7.1: Maps of the 95<sup>th</sup> percentile hourly concentration of  $SO_2$  at each flight level for NAME (left) and EMEP4UK (right).



Figure 7.2: Maps of the 95<sup>th</sup> percentile hourly concentration of SO<sub>4</sub> at each flight level for NAME (left) and EMEP4UK (right).



Figure 7.3: Maps of maximum hourly SO<sub>2</sub> concentration at each flight level for NAME (left) and EMEP4UK (right).



Figure 7.4: Maps of maximum hourly SO<sub>4</sub> concentration at each flight level for NAME (left) and EMEP4UK (right).

## 7.2 Probability of Exceeding Thresholds

The data produced by the models are informative about the probability of air quality thresholds being exceeded, but given the limitations of the study do not provide a full evaluation of the probability. Instead, the results demonstrate the relative frequencies of exceedance occurrence – i.e. the number of simulations in which a threshold was exceeded – rather than the probability of exceedance. These relative frequencies are discussed in this section, but for ease they are also referred to as probabilities.

The probability (relative frequency) of a threshold j being exceeded **at least once** at a particular flight level in a 6-week Laki-type eruption is given by the following equation for each grid box at that flight level:

$$PA_{j} = \frac{\sum_{i=1}^{80} E_{ij}}{80}$$

Where:

 $PA_j$  = the relative frequency of any time-step in a 6-week eruption exceeding a threshold j. This is evaluated from the 80 eruption periods considered.  $E_{ij}$  = 0 or 1 depending on whether the threshold j is exceeded at any time-step in eruption period i or not.

Maps of the relative frequency with which each threshold is exceeded in each flight level for each grid box are given in Annex C for hourly means. Values are given as a percentage. The lowest value possible in this calculation (other than zero) is 1 in 80 (i.e. in one simulation out of the 80 this threshold was exceeded at least once), which is 1.25%. For all locations and all flight levels, the probability of exceeding the Low1 lower threshold is 100%, as this threshold is set to zero. Tables 7.1 and 7.2 list the highest probability found in any grid square in the UK and Republic of Ireland region (Figure 7.5). These values are indicative only and are not necessarily representative of the whole region. The maps should be used to provide information on the spatial distribution.

These data show that in 100% of simulations the SO<sub>2</sub> threshold of Moderate4 is exceeded somewhere in the UK and Ireland region at FL240, FL300 and FL370. At lower flight levels the number of simulations where this is exceeded is lower, but still greater than 7%. The relative frequency of exceeding thresholds decreases with increasing AQI index number. However, the percentage of simulations in which SO<sub>2</sub> concentrations exceed the Very Hazardous12 threshold is still substantial at the upper flight levels; up to approximately 58% at FL300. Values drop off for the Very Hazardous13 threshold, with no level exceeding 5%. The percentages are greatest at the higher flight levels mainly because these correspond to the altitudes at which the most SO<sub>2</sub> is released from the volcano.

For SO<sub>4</sub> the maximum percentage of simulations that exceeded the Moderate4 threshold anywhere in the UK and Ireland region is approximately 89% for FL070 (Table 7.2). This drops to a maximum of ~6% at FL370. For the Very Hazardous12 threshold the maximum relative frequency of exceedance ranges from 0% to 45% depending on the flight level and model. The trend in probabilities for SO<sub>4</sub> is the reverse to SO<sub>2</sub>, with the highest percentages seen in the lowest flight levels. This is due to the time required for the conversion of SO<sub>2</sub> to SO<sub>4</sub>, and also suggests that the upper level plumes are moved over the UK too rapidly for a large SO<sub>4</sub> concentration

to have built up in the majority of cases. The elevated lower level concentrations could be due to both (a) atmospheric descent of plume and (b) lower level emissions of  $SO_2$  in certain eruption phases. Further investigation of the raw data would be needed to ascertain the contributions of these processes.

The maps for both species in Annex C show the influence of the dominant westerly flow over Iceland on the dispersion of the volcanic plumes. At FL150 and above percentages for both species are highest to the east of Iceland towards Norway. At FL070, there is more of a north-westerly influence and the distribution of the highest percentages is more towards the UK.

AQI	FL010	FL070	FL150	FL240	FL300	FL370
Level						
2	38.75	95.0	100	100	100	100
3	22.5	82.5	98.75	100	100	100
4	16.25	63.75	93.75	100	100	100
5	10.0	51.25	88.75	100	100	97.5
6	6.25	42.5	81.25	98.75	100	97.5
7	5.0	36.25	77.5	97.5	100	96.25
8	3.75	27.5	71.25	92.5	98.75	90.0
9	2.5	26.25	60.0	88.75	93.75	82.5
10	1.25	21.25	53.75	83.75	91.25	77.5
11	0	11.25	31.5	70.0	77.5	63.75
12	0	5.0	13.75	47.5	53.75	42.5
13	0	0	0	3.75	2.5	5.0

	FL010	FL070	FL150	FL240	FL300	FL370
2	30.0	95.0	100	100	100	100
3	13.75	73.75	98.75	100	100	100
4	7.5	56.25	95.0	100	100	100
5	5.0	43.75	92.5	100	100	100
6	2.5	38.75	88.75	97.5	100	100
7	1.25	31.25	86.25	96.25	100	100
8	1.25	21.25	78.75	87.5	100	97.5
9	0	17.5	75.0	76.25	97.5	95.0
10	0	13.75	70.0	68.75	96.25	92.5
11	0	5.0	47.5	46.25	85.0	77.5
12	0	1.25	20.0	16.25	57.5	46.25
13	0	0	0	1.25	3.75	5.0

Table 7.1: Maximum percentage relative frequencies of exceeding the lower threshold of each of the AQI bands for SO<sub>2</sub> simulated (using hourly mean output) at any time in a 6-week eruption period for a grid box anywhere over the UK and Ireland region derived from NAME (top) and EMEP4UK (bottom) model data. The Low1 band has been omitted as the lower threshold concentration is zero.

AQI	FL010	FL070	FL150	FL240	FL300	FL370
Level						
2	100	100	93.75	75.0	57.5	35.0
3	95.0	96.25	85.0	56.25	33.75	17.5
4	85.0	88.75	81.25	46.25	23.75	6.25
5	78.75	85.0	75.0	42.5	20.0	2.5
6	72.5	80.0	71.25	40.0	18.75	1.25
7	66.25	76.25	70.0	36.25	17.5	1.25
8	65.0	72.5	68.75	36.25	15.0	1.25
9	57.5	70.0	67.5	33.75	13.75	1.25
10	53.75	66.25	65.0	28.75	12.5	1.25
11	27.5	51.25	53.75	17.5	6.25	0
12	21.25	45.0	45.0	10.0	3.75	0
13	11.25	31.25	37.5	6.25	2.5	0

AQI	FL010	FL070	FL150	FL240	FL300	FL370
Level						
2	90.0	82.5	92.5	100.0	81.25	23.75
3	62.5	53.75	56.25	96.25	46.25	7.5
4	48.75	32.5	30.0	76.25	23.75	3.75
5	42.5	30.0	17.5	60.0	21.25	2.5
6	38.75	22.5	12.5	50.0	17.5	2.5
7	33.75	21.25	10.0	42.5	16.25	1.25
8	28.75	16.25	8.75	37.5	15.0	1.25
9	25.0	13.75	7.5	33.75	10.0	1.25
10	22.5	11.25	5.0	30.0	10.0	1.25
11	3.75	0	0	6.25	2.5	0
12	1.25	0	0	1.25	0	0
13	0	0	0	0	0	0

Table 7.2: Maximum percentage relative frequencies of exceeding the lower threshold of each of the AQI bands for SO<sub>4</sub> simulated (using hourly mean output) at any time in a 6-week eruption period for a grid box anywhere over the UK and Ireland region derived from NAME (top) and EMEP4UK (bottom) model data. The Low1 band has been omitted as the lower threshold concentration is zero.



Figure 7.5: The green outline depicts the area evaluated as the UK and Ireland region for the tables in Section 7.

### 7.3 Consecutive Duration of Exceedances

The consecutive length of time that each grid box remains above each of the Air Quality thresholds from the Moderate4 band upwards has been calculated at each flight level for a domain around the UK for all of the 80 simulations. The maximum consecutive duration in any of the runs has then been extracted for each grid square. This gives an indication of the "worst-case" duration of exposure that could be expected. Note that during one 6-week eruption period there could be multiple instances of concentrations remaining above a certain threshold. For context, the total number of hours modelled by each model was 80640 hours, of which 67200 were during emission periods.

Maps of the maximum number of consecutive hours of exceedance of a certain threshold at the each flight level are given in Annex C. This is the maximum consecutive duration modelled in each grid box in any of the 80 simulated periods. Tables 7.3 and 7.4 list the maxima of these maximums (i.e. the highest consecutive duration simulated in any grid box in the domain shown in the Annex C maps). Note: these values are the time spent at and above the AQI level lower threshold, not the time spent within each AQI band. Values in the tables are not necessarily for the same grid box at each flight level.

These data reveal that although the probability of  $SO_2$  thresholds being exceeded at the lower flight levels is less than for the upper levels, when they are exceeded episodes can still last for many hours (up to 30 hours for the Moderate4 threshold at FL010). The longest consecutive durations occur at FL370, with a maximum consecutive length of exceedance of the  $SO_2$  Moderate4 threshold of approximately 5 days.

For SO<sub>4</sub>, the maximum consecutive length of exceedance of the SO<sub>4</sub> Moderate4 threshold is less than a day at FL370, but is over 5 days at FL010. This longer duration of consecutive events in the lower flight levels for SO<sub>4</sub> compared to SO<sub>2</sub> is consistent with the findings from the other statistics.

For exceedances of the Hazardous11 thresholds, maximum consecutive durations are longer than a day at all flight levels except FL010 and FL070 for  $SO_2$ , and at the three lower levels for  $SO_4$ .

The figures in Annex C are very noisy and show traces of many individual plume features at most levels and all thresholds. There is a particularly obvious cross pattern in Figure C-20 for example. That individual features are apparent implies that the use of 80 simulations does not give an extensive enough dataset for robust analysis of the maximum consecutive duration at these flight levels. It also prevents detailed analysis of any trends in the geographical distribution of the longer durations, beyond the observation that durations of SO<sub>2</sub> above thresholds appear longer closer to lceland for the upper flight levels.

AQI	FL010	FL070	FL150	FL240	FL300	FL370
Level						
4	30	56	66	73	61	111
5	22	55	62	63	61	109
6	18	53	56	56	60	96
7	15	40	54	53	59	95
8	11	36	49	45	57	94
9	7	34	48	43	57	94
10	4	29	47	40	55	92
11	0	17	42	32	45	54
12	0	10	18	26	23	32
13	0	2	2	4	9	10

AQI	FL010	FL070	FL150	FL240	FL300	FL370
Level						
4	29	99	72	57	97	104
5	17	97	70	50	90	103
6	11	95	57	46	84	78
7	7	93	56	42	60	78
8	2	72	54	38	53	65
9	0	47	54	33	53	59
10	0	40	53	30	52	57
11	0	23	45	25	41	53
12	0	6	9	18	33	27
13	0	0	0	2	5	11

Table 7.3: Maximum consecutive hours above  $SO_2$  thresholds in any grid box in the region depicted in the maps in Annex C from NAME (top) and EMEP4UK (bottom) model data.

AQI	FL010	FL070	FL150	FL240	FL300	FL370
Level						
4	129	99	66	55	24	16
5	120	96	65	41	22	16
6	118	95	64	36	22	10
7	115	94	61	31	22	7
8	115	94	60	30	22	6
9	112	87	60	30	22	5
10	112	84	58	30	22	3
11	66	77	43	21	17	0
12	58	55	36	19	14	0
13	52	42	28	11	9	0

AQI	FL010	FL070	FL150	FL240	FL300	FL370
Level						
4	122	83	37	52	33	8
5	109	77	30	50	22	6
6	102	65	24	47	21	5
7	100	63	20	47	20	5
8	99	61	19	46	18	4
9	99	59	18	41	16	3
10	97	56	17	38	15	2
11	38	0	0	12	6	0
12	5	0	0	3	0	0
13	0	0	0	0	0	0

Table 7.4: Maximum consecutive hours above SO<sub>4</sub> thresholds in any grid box in the region depicted in the maps in Annex C from NAME (top) and EMEP4UK (bottom) model data.

#### 7.4 Mean Total Time Thresholds are Exceeded

The mean number of hours a certain threshold is exceeded at each flight level in a 6week eruption has been derived from the total number of hours where the threshold was exceeded across of all the 80 simulation periods. The following equation has been applied to each modelled grid box for each flight level:

$$\overline{T}_j = \frac{\sum_{i=1}^{80} T_{ij}}{80}$$

Where:

 $\overline{T}_{j}$  = the mean number of hours above a threshold j in a 6-week eruption period for one flight level

 $T_{ii}$  = the total number of time-steps above threshold j in eruption period i

Maps of the calculated mean number of hours that a certain threshold is exceeded in a 6-week eruption period are given in Annex C for each flight level. These give an indication of the mean total time a threshold could be exceeded during an eruption. As these values are a mean, they will be considerably shorter than the maximum consecutive durations in Section 7.3. Only mean values above 1 hour are plotted.

Tables 7.5 and 7.6 list the highest mean total-time seen in any of the grid boxes over the UK and Ireland. This value may only occur in one grid box. Values in the tables are not necessarily for the same grid box at each flight level or AQI level. Also note that these values are the time spent at and above the AQI level lower threshold, not the time spent within each AQI band. These values are indicative only and are not necessarily representative of the whole region. The maps should be used to provide information on the spatial distribution.

These data reveal that at FL010 there is no exceedance of 1 hour or longer of the Moderate4 AQI threshold of 267  $\mu$ g/m<sup>3</sup> over the UK. Across all of the flight levels, the total threshold exceedance times for SO<sub>2</sub> are generally highest in the near-field of the volcanic vent and there is a gradual decrease in the total exceedance time with distance from the volcanic vent. At FL370 there is good agreement between EMEP4UK and NAME, with NAME predicting that the Moderate4 SO<sub>2</sub> threshold is exceeded for up to 495 hours in the near-field and EMEP4UK predicting 537 hours. For comparison at FL370 over the UK and Ireland the maximum total exceedance time is approximately 28 hours.

In general, for SO<sub>4</sub> there is a very low number of hours (below 24) during which the Moderate4 threshold of 36  $\mu$ g/m<sup>3</sup> (or any other value) is exceeded in the far-field in any flight level. The High threshold of 54  $\mu$ g/m<sup>3</sup> is exceeded only in FL010 to FL240 in the far-field for between 2 and 14 hours.

AQI	FL010	FL070	FL150	FL240	FL300	FL370
Level						
4	<1	5.8	17.5	26.8	28.9	27.6
5	<1	4.1	12.8	20.7	23.5	22.5
6	<1	3.1	9.6	16.8	19.6	18.7
7	<1	2.7	7.5	13.6	16.4	16.0
8	<1	2.0	4.8	9.7	12.0	11.8
9	<1	1.5	3.3	7.2	9.1	9.1
10	<1	1.1	2.6	5.6	7.5	7.5
11	0	<1	1.3	3.3	4.5	4.2
12	0	<1	<1	1.5	1.8	1.9
13	0	0	0	<1	<1	<1

Table 7.5: Maximum mean total time (hours >1) above  $SO_2$  thresholds simulated in a grid box anywhere over the UK and Ireland at the flight level specified derived from NAME model data.

AQI	FL010	FL070	FL150	FL240	FL300	FL370
Level						
4	23.2	18.4	10.4	3.4	1.4	<1
5	19.5	15.6	9.2	2.7	1.2	<1
6	16.6	13.8	8.3	2.3	<1	<1
7	14.1	12.4	7.7	2.0	<1	<1
8	12.5	11.6	7.3	1.8	<1	<1
9	10.8	10.7	6.8	1.6	<1	<1
10	9.4	9.9	6.5	1.4	<1	<1
11	3.9	6.0	4.3	<1	<1	0
12	2.5	4.2	3.3	<1	<1	0
13	1.5	2.4	2.1	<1	<1	0

Table 7.6: Maximum mean total time (hours >1) above  $SO_4$  thresholds simulated in a grid box anywhere over the UK and Ireland at the flight level specified derived from NAME model data.

### 7.5 Arrival Times

Seven regional boxes have been constructed to represent the UK and Republic of Ireland (Fig 7.6). A further three boxes have been specified to cover core airports in the UK. The south-east is of particular interest as any disruption in this area would affect the bulk of UK long-haul air travel for which there are no alternative transport options, for this reason the box has been chosen to cover the four major London airports (Heathrow, Gatwick, Stanstead, Luton). These boxes have been used to determine the fastest arrival time of a plume after the start of the eruption in each of the 80 scenarios. Note that the arrival times of plumes from different eruptive episodes during the eruption scenario cannot be easily calculated and could be faster.

Using NAME data the quickest time (in hours) of arrival of a plume to each of the boxes has been calculated. The mean time of arrival has also been calculated from all of the 80 scenarios modelled. For flight altitudes the arrival times refer to when the Moderate4 threshold for  $SO_2$  is exceeded (>267 µg/m<sup>3</sup>). This threshold has been used at the specification of the CAA. The mean time is calculated from only those periods when a plume above the Moderate4 threshold reached the box.



Figure 7.6: The seven regions (red) and airport zones (light blue) chosen for the arrival time analysis.

Table 7.7 shows the quickest arrival time of the  $SO_2$  plume to each box. Table 7.8 shows the mean arrival time of  $SO_2$ , as well as the percentage of the 80 simulations where the Moderate4 threshold level was exceeded at each box and which were used in the mean calculation. This demonstrates that the plume arrives much faster in the upper flight layers, where the majority of the emissions are occurring and where wind speeds are higher on average. The fastest arrival time to Scotland at these altitudes is only 6-7 hours, but the whole of the UK can be reached within 24 hours. Case study 1 looks at one of these fast arrival episodes in more detail. The

mean arrival time over the UK at the higher altitudes is on the order of one week, although at certain flight levels it is only 4 days for Scotland.

At the lower levels where little material is directly emitted and the gas plume has to be brought down by meteorological systems to reach the elevated concentrations, the arrival times are considerably longer. Exceedances of the Moderate4 threshold near the surface are not common, hence the arrival times are more uncertain at the lowest levels. (Note: when comparing to the equivalent tables in Section 6, it is important to remember the different thresholds considered).

Box	Location	FL010	FL070	FL150	FL240	FL300	FL370
1	Northern	375	155	20	10	9	17
	Ireland	(15.6)	(6.5)				
2	Republic of	375	59	12	7	6	7
	Ireland	(15.6)	(2.5)				
3	Scotland	378	36	11	7	6	7
		(15.8)	(1.5)				
4	Wales	379	71	33	12	10	17
		(15.8)	(3)				
5	Northern	400	84	14	10	9	10
	England	(16.7)	(3.5)				
6	Central & East	402	54	17	12	11	13
	England	(16.8)	(2.25)				
7	Southern	385	71	20	14	12	17
	England	(16)	(3)				
8	London	413	103	19	14	13	15
	Airports	(17.2)	(4.3)				
9	Manchester	452	189	46	12	11	13
	Airport	(18.8)	(7.9)				
10	Glasgow &	685	160	14	9	9	10
	Edinburgh	(28.5)	(6.7)				

Table 7.7: Quickest arrival time of SO<sub>2</sub> (>267  $\mu$ g/m<sup>3</sup>) in hours and (days) to each box and altitude.

Box	Location	FL010	FL070	FL150	FL240	FL300	FL370
1	Northern	620	530	252	174	164	204
	Ireland	(25.8)	(22.1)	(10.5)	(7.3)	(6.8)	(8.5)
		12.5%	66.25%	92.5%	97.5%	97.5%	91.25%
2	Republic of	618	484	270	172	164	224
	Ireland	(25.8)	(20.2)	(11.3)	(7.2)	(6.8)	(9.3)
		18.75%	72.5%	93.75%	98.75%	98.75%	93.75%
3	Scotland	675	423	193	103	106	145
		(28.1)	(17.6)	(8.0)	(4.3)	(4.4)	(6.0)
		23.75%	87.5%	100%	100%	100%	100%
4	Wales	580	548	324	199	177	206
		(24.2)	(22.8)	(13.5)	(8.3)	(7.4)	(8.6)
		15%	71.25%	91.25%	100%	98.75%	86.25%
5	Northern	700	509	262	162	153	184
	England	(29.2)	(21.2)	(10.9)	(6.8)	(6.4)	(7.7)
		22.5%	75%	96.25%	100%	100%	97.5%
6	Central &	724	542	274	160	162	190
	Eastern	(30.2)	(22.6)	(11.4)	(6.7)	(6.8)	(7.9)
	England	20%	75%	93.75%	98.75%	98.75%	91.25%
7	Southern	653	505	314	172	170	206
	England	(27.2)	(21.0)	(13.1)	(7.2)	(7.1)	(8.6)
		20%	71.25%	90%	100%	100%	92.5%
8	London	735	574	380	206	210	235
	Airports	(30.6)	(23.9)	(15.8)	(8.6)	(8.8)	(9.8)
		11.25%	47.5%	78.75%	97.5%	98.75%	82.5%
9	Manchester	735	584	373	242	201	212
	Airport	(30.6)	(24.3)	(15.5)	(10.1)	(8.4)	(8.8)
		8.75%	50%	80%	98.75%	97.5%	82.5%
10	Glasgow &	751	585	286	181	181	181
	Edinburgh	(31.3)	(24.4)	(11.9)	(7.5)	(7.5)	(7.5)
	Airports	8.75%	56.25%	90%	100%	100%	96.25%

Table 7.8: Mean arrival time of SO<sub>2</sub> (>267  $\mu$ g/m<sup>3</sup>) in hours and (days), and the percentage of simulations in which the Moderate4 threshold was exceeded for each box and altitude.

# 7.6 Case-studies of high impact periods

A huge amount of data has been produced in this work and the summary results presented so far provide no detail on specific periods or the evolution of pollution episodes. Analysis of the raw hourly outputs from the models shows that the plumes can be highly transient in nature and their location is strongly controlled by the meteorology. To provide more detail on the time evolution of the SO<sub>2</sub> and SO<sub>4</sub> plumes over the UK, a selection of case studies are presented. These have been derived from the NAME model data. Due to the time-varying natures of the plumes, identifying suitable case studies is more challenging for the aviation levels than the near-surface.

# 7.6.1 Case-Study 1: Rapid transport of SO<sub>2</sub> to the UK at upper flight levels

The fastest travel time of an SO<sub>2</sub> plume to London airports occurs at the start of simulation Period 28. In this case, the Moderate threshold ( $267 \mu g/m^3$ ) is exceeded in the upper flight levels (FL240, FL300, FL370) within 15 hours of the start of the eruption. The emissions relate to those in the first part of day 1 of the eruption, which is the 30 May 2006. On this day the bulk of the volcanic SO<sub>2</sub> is emitted between 6 and 12 km asl above the volcanic vent, which corresponds to the affected flight levels. The rapid transport of the plume over such a large distance from Iceland (>1800 km) has the consequence of keeping the plume very compact and with a small lateral

spread, so that a relatively narrow horizontal area is affected at each of the flight levels (Fig 7.7). This means that high concentrations are maintained in a relatively narrow plume. The plume is also very transient in its location and quickly moves away to the west. This behaviour would appear to be closely related to the location of the jet stream, which was over the UK at this time (Fig 7.8).



Figure 7.7: Rapid transport of the SO<sub>2</sub> plume to the UK on day 1 of Period 28 at FL370.



Figure 7.8: Winds speeds (in m/s) at 12 km asl at 12:00 UTC on 30 May 2006, showing the position of the jet stream over the North Atlantic and UK and its proximity to Iceland.

# 7.6.2 Case-Study 2: Variable timing and concentrations at different levels

Section 7.5 showed that the time of arrival of a plume to the UK varies with altitude. This case study demonstrates that during times of north westerly flow from Iceland different flight levels can be affected by volcanic plumes on different days, resulting in a complicated four-dimensional situation for the air above the UK. On the 12-14 May 2003 (simulation Period 4) the transport and dispersion of the simulated volcanic SO<sub>2</sub> plume from Iceland is primarily south-eastwards directly towards the UK. The resulting SO<sub>2</sub> concentrations in the upper four flight levels are shown in Figures 7.9-7.12. The situation in each of these is different and summarised as follows:

- At FL150, a plume with VeryHigh+ concentrations of SO<sub>2</sub> reaches the UK on the morning of the 14 May, with Northern Ireland, Wales and the south-west of England experiencing the highest concentrations.
- At FL240, a Moderate concentration plume crosses Wales and central England from west to east during the second half of the 13 May. Subsequently a plume with Hazardous+ concentrations of SO<sub>2</sub> reaches the UK on the afternoon of the 13 May, 6-12 hours earlier than that at FL150, and at 00:00 on the 14 May is affecting a large north-south zone of the UK.
- At FL300, the UK is affected by up to Very Hazardous12 levels of SO<sub>2</sub> throughout the 12, 13 and 14 May, with larger areas affected in the earlier stages, as the plume initially moves over the UK to the east.
- At FL370, a plume with Very Hazardous13 concentrations of SO<sub>2</sub> is emitted from the volcano and this concentration is sustained as the plume is transported rapidly to the UK (approximately 24 hours to reach Scotland) and then proceeds to cross Scotland and Northern England. This plume is responsible for the black, Very Hazardous13, swath seen over the UK in the NAME maximum plot for FL370 in Figure 3.

From a UK perspective, the location of the plume at FL370 at 00:00 on 14 May is in a very different to the location of the plume in the other flight levels, with the consequence that almost the whole of the country is covered by a plume at one level or another. This example shows that the situation at one flight level cannot be extrapolated to represent other levels in this data set, either in terms of timing, location or concentration. It also demonstrates that knowing the height of the emission of the SO<sub>2</sub> from a volcano can be critical in determining where and when might be affected.

For SO<sub>4</sub> the situation is quite different, with concentrations at FL240, FL300 and FL370 never exceeding the Low AQI bands. At FL150, Moderate4 is only exceeded in a very small area of Wales/the Midlands in the morning of the 13 May. This demonstrates the important point that plumes with elevated levels for SO<sub>2</sub> and for SO<sub>4</sub> do not necessarily coincide in time or space.



Figure 7.9: Location and concentration of SO<sub>2</sub> plume at FL150 on 12-14 May 2003.



Figure 7.10: Location and concentration of SO<sub>2</sub> plume at FL240 on 12-14May 2003.



Figure 7.11: Location and concentration of SO<sub>2</sub> plume at FL300 on 12-14May 2003.



Figure 7.12: Location and concentration of SO<sub>2</sub> plume at FL350 on 12-14May 2003.

# 8 Assessment of Uncertainty

There is considerable uncertainty in the source term for modelling an effusive eruption of the scale of the 1783 Laki eruption. To address this uncertainty fully was beyond the scope of the funded project. Instead, to provide a limited assessment of the potential range of uncertainty, two approaches have been followed:

- 1. A number of simulations have been conducted for one of the periods used in the initial modelling study conducted in 2013 by the Met Office and University of Leeds for the CSS H55 Project Board.
- 2. A stochastic Monte Carlo assessment has been conducted for the case study period in Section 6.7.2

The results provide an indication of the scale of uncertainty that needs to be considered when analysing the main results from the project in Sections 6 and 7 and will hopefully provide some context to the Stakeholder Department impact assessments.

### 8.1 Method 1: Sensitivity tests

An initial modelling study was conducted in 2013 by Claire Witham (Met Office), Matthew Hort (Met Office) and Anja Schmidt (University of Leeds) using the NAME model. This considered 11 short eruption periods from April to August 2010, where each eruption lasted only 6-hours and the related emissions were followed for 14 days. This time period was deliberately chosen so it contained the Eyjafjallajokull eruption period, which included significant northerly flow from Iceland. The initial work demonstrated a surface hazard to Europe from sulphur dioxide and sulphate aerosol, with the period of 12:00 UTC 23/06/2010 to 12:00 UTC 7/7/2010 resulting in "very high" (on the UK Air Quality Index) concentrations of sulphur dioxide (SO<sub>2</sub>) and sulphate aerosol (SO<sub>4</sub>) near the surface over Europe (Figure 1.1). This was the only period in the eleven modelled where significantly elevated concentrations of SO<sub>2</sub> were simulated near the surface.

To test how these concentrations vary when certain limited changes are made to the source term, six new NAME model runs for the identified 2-week period were conducted. These test the impact of:

- Increasing and decreasing the mass of SO<sub>2</sub> emitted.
- Increasing and decreasing the release height of the emitted SO<sub>2.</sub>
- Changing the duration of the emission.

Table 8.1 outlines the differences applied to the source terms in each model run compared to the initial simulation. The naming of the scenario runs indicates the changes made in the set-up: M represents a change in the mass emitted, H a change in the height, and T a change in the duration. The initial scenario had an emission of 10 Mt of SO<sub>2</sub> over 6-hours. This was emitted uniformly between 5-10 km asl. The transport and conversion of this emission over the next 14 days was then simulated.

It is important to note that the single pulse of  $SO_2$  that is emitted in these simulations is different to the emissions used in the rest of the modelling project, which vary vertically and temporally. The findings should translate to a more complex source term however.

	Height Range (km asl)							
SO₂ Mass (Tg)	Surface-5		5-10			10-15		
5			M2					
10	H2		Initial	T1	T2	H1		
20			M1					

Table 8.1: Eruption source parameters used for each model run. Light, medium and dark shading indicate release durations of 6, 12 and 24 hours respectively. Model identifiers indicate which combination of parameters has been considered here. Note: 1Tg = 1 Mt.

#### 8.1.1 Model set-up specifics

Only the NAME model has been used for these sensitivity tests. The model set-up was:

- Emissions were released from a 100 m diameter column above the location 64.064N, 18.22W, which is in the area of the Laki fissures from 1783.
- Simulations were conducted on a domain covering 25N to 80N and 45W to 45E.
- NAME's chemistry scheme was used. The setup option of "ozone on the background field" was selected. Anthropogenic emissions were not included.
- Wet and dry deposition were allowed.
- Numerical Weather Prediction (NWP) data was from the Met Office's global MetUM model at a horizontal resolution of approximately 25 km.
- All emissions started at 12:00 UTC 23/06/2010.
- For each emission scenario the simulations were run for 14 days.
- Outputs were produced on a horizontal grid of resolution 0.5 x 0.5 degrees latitude-longitude.
- Concentration values have been output every hour and are the average for the 60 minutes up to the stated time. SO<sub>2</sub> and SO<sub>4</sub> are available for the near-surface and SO<sub>2</sub> is available at altitude.
- Near-surface concentrations are a model-calculated boundary-layer average value. (These SO<sub>2</sub> concentrations are output by the model as mass mixing ratios and are presented here in μg/m<sup>3</sup>. Conversion to ppb is possible).
- No account is made for existing background levels of pollution in the maps and the values presented here.
- The level FL300-FL325 (approximately 9000-11000 m above sea level) has been selected to assess the impact at cruising flight altitudes, and is here referred to as FL300

#### 8.1.2 Results

#### 8.1.2.1 Near-surface

Maps of the maximum hourly near-surface concentrations of  $SO_2$  and  $SO_4$  are contained in Annex D for each simulation together with hourly illustrative snapshots for the same "worst-case" times as Figure 8.1. These show the differences in distribution of the pollutants resulting from each of the scenarios. In each case, apart from H1, High and Very High levels of  $SO_2$  and  $SO_4$  are simulated near the surface over Germany and Italy. These levels start to appear about 3 days after the start of the emission for  $SO_2$  and 4 days after for  $SO_4$  in the Initial, M1 and M2 simulations. For T1 and T2 the arrival of both  $SO_2$  and  $SO_4$  occurs simultaneously on about day 4. H2 is the only scenario where the  $SO_4$  cloud arrives first, after about 3 days, having been created and transported in the lower atmosphere. High levels of  $SO_2$  are only seen on day 4 and later in this scenario.



Figure 8.1: Example "worst-case" hourly boundary-layer levels simulated in the initial modelling of this period for (a) sulphur dioxide (at 18:00UTC 27/06/2010) and (b) sulphate aerosol (at 03:00 01/07/2010) plotted on the UK Air Quality Index scales.

Table 8.2 lists the maximum value recorded in any one-hour time period for both  $SO_2$  and sulphate for each model scenario. The differences between the values are perhaps more informative than the numbers themselves for understanding uncertainty. Table 4 reveals that changing the mass emitted results in concentrations of  $SO_2$  that differ by the same factor as the initial mass release, i.e. the variability in the M1 and M2 scenarios is directly proportional to the initial model emission. This indicates that the conversion of  $SO_2$  is acting at the same rate in the Initial, M1 and M2 scenarios and that any other loss processes, e.g. wet deposition, are acting linearly. The situation is more complex for  $SO_4$  however.

No such linkages can be made for the H1 and H2 concentrations as, unlike the mass emitted, the height of the emission is not directly linked to the concentration. Rather the height of the emission affects the transport and whether material is brought to the

ground, so for the high-altitude emission scenario H2 very little of either species reaches the surface. In T1 and T2 the maximum concentrations are not too dissimilar to the Initial 6-hour release. More detailed investigation is needed to explore the reason(s) for this.

Table 8.2 demonstrates that the maximum concentrations experienced can vary by approximately an order of magnitude (i.e. a factor of 10) for  $SO_2$  depending on emission heights and mass (excluding H1). For  $SO_4$  the range is much greater, around a factor of 30 (excluding H1). Changing the duration of emission has a smaller impact in this case than changing the height or mass, with maximum  $SO_2$  concentrations varying by only approximately 15%. For  $SO_4$  the values vary within a factor of two.

SO <sub>2</sub> Mass		Height Range (km asl)								
(Tg)		Surface-5		5-10			10-15			
F	SO <sub>2</sub>				1470					
5	SO <sub>4</sub>				161					
10	SO <sub>2</sub>	660			2890	3010	2520	11		
10	SO <sub>4</sub>	1180			963	1670	1010	26		
20	SO <sub>2</sub>				5660					
20	SO <sub>4</sub>				5090					

Table 8.2: Maximum near-surface hourly SO<sub>2</sub> and SO<sub>4</sub> modelled concentrations over mainland Europe and the Mediterranean region in each simulation. Light, medium and dark shading indicate release duration of 6, 12 and 24 hours.

To give a better representation of the spread of maximum hourly concentrations, Figures 8.2 and 8.3 show the maximum hourly value recorded for each grid square (including those over the sea) that experienced a maximum over AQ index threshold 4 (i.e.  $266 \mu g/m^3$  for SO<sub>2</sub> and  $35 \mu g/m^3$  for SO<sub>4</sub>). Note that these maximum values do not necessarily occur at the same time for each grid square. The "area affected" value has been calculated by assuming that each grid square is approximately 55km by 35km in size (i.e. an approximation of the 0.5 x 0.5 degree output grid at European latitudes). For H2, the low emission height means that Iceland is impacted by very high concentrations of both species immediately following the emission (Figure 8.4). This explains many of the High and Very High values seen for H2 in Figures 8.2 and 8.3. H1 is not shown as the upper altitude emission results in minimal material reaching the surface and index 4 is never reached.

Figures 8.2 and 8.3 show considerable variability between the simulations. For SO<sub>2</sub>, the M1 scenario with double the emitted mass results in the largest areas being affected. This is due to the relative increase in concentrations at each grid square. The longer duration emission scenarios also result in larger areas being affected by moderate and high levels compared to the initial run. This is due instead to the role of the weather in transporting the later emissions to different locations. In general for SO<sub>2</sub> the area affected decreases for higher AQ index bands. For SO<sub>4</sub> however all simulations show a significant peak in the area affected for the Very High and Very High+ categories. This demonstrates that using the daily AQ index scale for PM2.5 for assessing the distribution of the sulphate values is potentially unhelpful, as it does not offer a graduation above 70  $\mu$ g/m<sup>3</sup> and consequently does not aid understanding of which areas experience the highest concentrations.

Analysis of the values for each threshold in the figures demonstrates that a reasonable estimate of the uncertainty on the area affected at each concentration threshold is:



Figure 8.2: Distribution of the maximum SO<sub>2</sub> concentrations simulated for every one of the grid squares, binned by AQ index threshold from Moderate index 4 upwards.



Figure 8.3: Distribution of the maximum SO<sub>4</sub> concentrations simulated for every one of the grid squares, binned by AQ index threshold from Moderate index 4 upwards. The Very High category contains values from 70-700  $\mu$ g/m<sup>3</sup> and the Very High+ category contains all values greater than 700  $\mu$ g/m<sup>3</sup>.

a factor of three for  $SO_2$ 

• a factor of 1.5 for SO<sub>4</sub> for index levels up to Very High



Figure 8.4: Near-surface SO<sub>2</sub> concentrations 24 hours after the start of emission in simulation H2. In all other simulations there are no elevated concentrations anywhere in the domain at this time.

It is also of interest to consider the length of time that levels remain above certain thresholds. Tables 8.3 and 8.4 show the maximum episode length (in hours) when concentrations are maintained at and above the levels listed for *anywhere* in the model domain. Note, when levels are above High they are also counted as being above Moderate, and the values similarly cascade for the Very High category. This is not the same as the maximum duration at an individual grid square, but given the nature of this episode and its persistence over Germany the Very High data could be taken as being broadly representative of individual grid squares in this region for SO<sub>2</sub>. The longer persistence of SO<sub>4</sub> is related to the southward movement of the plume over time, so the same relationship to individual grid squares cannot be assumed. The lifetime of SO<sub>4</sub> means that in many cases levels were still elevated over Very High somewhere in the model area at the end of the simulation, this is indicated by a plus symbol "+" in the data column.

SO <sub>2</sub> Mass	Index		Height Range (km asl)							
(Tg)	Value	Surface-5			5-10			10-15		
	≥ 4				80					
5	≥ 7				67					
	≥ 10				16					
	≥ 4	51			167	167	145	0		
10	≥ 7	17			97	78	90	0		
	≥ 10	0			67	90		0		
	≥ 4				211					
20	≥ 7				167					
	≥ 10				77					

The results in Tables 8.3 and 8.4 show that episodes of Very High  $SO_2$  and  $SO_4$  have the potential to persist for many days in all of the scenarios. For the lower concentration thresholds, the duration of the episodes are even longer.

Table 8.3: Maximum duration in hours at which any near-surface point in the model domain is at or above the AQ index level stated for SO<sub>2</sub>. H2 (Surface-5) values exclude the Iceland area.
SO <sub>2</sub> Mass	Index	Height Range (km asl)								
(Tg)	Value	S	urface	-5		5-10			10-15	
	≥ 4				242+					
5	≥ 7				212					
	≥ 10				167					
	≥ 4	258			262+	255+	242+	0		
10	≥ 7	240			242+	240	235	0		
	≥ 10	216			241+	220		0		
	≥ 4				264+					
20	≥ 7				264+					
	≥ 10				264+					

Table 8.4: Maximum duration in hours at which any near-surface point in the model domain is at or above the level stated for SO<sub>4</sub>. H2 (Surface-5) values exclude the Iceland area.

#### Altitude

Maps showing snap-shots of the development of the  $SO_2$  cloud at commercial aviation cruise altitudes (here chosen to be FL300-FL325) are given in Annex D. Because the  $SO_2$  is emitted directly into these altitudes, except in the low emission height scenario H2, the highest concentrations occur during the initial period of the cloud's development, unlike the near-surface results. To demonstrate the cloud's evolution five time-steps at 12-hourly intervals over the first three days are provided. These show that at FL300 the cloud is initially transported to the south towards and over northern Scotland (in many respects similar to the path of the Grimsvotn 2011 ash cloud). It is then transported north-eastwards along the coast of Norway, predominantly over the sea, and disperses as it travels. At lower altitudes (data not shown) the cloud shows a similar behaviour initially, but is then transported over Europe before elongating in a north-east south-west direction over central Europe and Scandinavia.

The maximum  $SO_2$  concentrations simulated at each of the five time-steps for FL300 are given in Table 8.5. The decrease in maximum concentration with time is related to both the gradual dispersion of the cloud and the chemical conversion of the  $SO_2$  to  $SO_4$ . As with the near-surface results, the altitude results show a clear linear relationship between the M1 and M2 values and the initial values, which is directly related to the difference in the mass emitted. No values are simulated at this altitude for H2 (all of the emission occurs below FL300), although upwards transport from lower emission heights is possible. For T1 and T2 the large maximums at T+12 (and T+24 for T2) are due to material still being emitted directly from the vent at these times, so these values reflect the levels at the vent rather than in the downwind cloud.

Time	Initial	M1	M2	H1	H2	T1	T2
T+12	16000	32100	8030	8740	0	48500	24400
T+24	11900	23800	6110	5050	0	6110	12700
T+36	12200	25000	6210	1590	0	9830	5200
T+48	10000	19900	4980	1070	0	8010	4000
T+60	6770	13200	3330	534	0	5480	2790

Table 8.5: Maximum concentration of SO<sub>2</sub> (ppb – note the change in units) at FL300 in the modelled cloud in each simulation for five time-steps after the start of the emission release. T+12 indicates 12 hours after the start and so on. Grey cells indicate values that are affected by continuing emissions from the source in Iceland.

## 8.1.3 Discussion

The six scenarios show a range of differences compared to the Initial model run. These provide some insight into the uncertainty that could be expected in the full modelling study results.

In the Initial model run, the weather plays a leading order role in the occurrence of Very High concentrations at the surface. During the simulation period high pressure is dominant over central Europe (Figure 8.5) leading to the descent of air, and any pollutants contained within it, towards the surface. Slack winds related to the high pressure mean that the air is not dispersed and pollution levels are maintained or increase as the air becomes trapped near the surface. When these types of conditions persist for a few days in the summer months (as they do during the modelled period) they are commonly associated with haze and decreased air quality. At altitude, the movement of the plume to the south over Scotland and then subsequently north-eastwards is related to the passage of a low pressure system from the North Atlantic to the north-east during this time.



Figure 8.5: Synoptic meteorology chart for 12:00 UTC on 1 July 2010 showing the dominant high pressure across central Europe and the Mediterranean. The lack of isobars in this region is indicative of slack surface winds.

The results from changing the mass emitted are the easiest to understand:

- In M1 the increased mass emission results in increases in SO<sub>2</sub> and SO<sub>4</sub> concentration.
- In M2 the decreased mass emission results in decreases in SO<sub>2</sub> and SO<sub>4</sub> concentration

For both M1 and M2 under these meteorological conditions, near-surface  $SO_2$  concentration appears to be linearly related to the emission rate, i.e. doubling the mass doubles the downwind concentrations, but for  $SO_4$  it is non-linear. Further investigation of these relationships is necessary as they may not hold in other emission scenarios and/or meteorological conditions. Increasing the mass emitted is the dominant factor for expanding the area affected at all concentration levels compared to the initial run.

At FL300, M1 and M2 exhibit the same cloud shape and dispersion as the initial run, but with different  $SO_2$  concentration values. Similar to the near-surface values the M1 values are double the initial values and the M2 values are half the initial values. This suggests that uncertainty in  $SO_2$  concentrations downwind at all altitudes can be directly linked to uncertainty in the mass emitted. A XX% uncertainty in the mass emitted as determined by expert elicitation will result in an XX% uncertainty in the modelled  $SO_2$  values if all other parameters are well constrained.

The height of the emission has the strongest influence (in these scenarios) on where the  $SO_2$  and  $SO_4$  clouds occur both in the horizontal and the vertical. In H1 the injection is predominantly above the tropopause - this is the boundary between the troposphere and the stratosphere and above Iceland is typically at around 10 km and so in this case the emissions do not get caught up in the tropospheric weather pattern that brings the material to the surface in the Initial scenario. This is an important point, as in general material injected into the stratosphere will predominantly have a minimal surface influence, due to lack of mixing between the troposphere and the stratosphere. There is still a hazard at aviation cruising levels however and the SO<sub>2</sub> cloud at FL300 in H1 has similar transport characteristics to that in the Initial scenario. The extent and concentration of the cloud are reduced in H1, which is related to the differing height of the emissions as the FL300 layer is probably only capturing the lowest extent of the plume in this scenario. If quantities are sufficient then there is more potential for a climate influence from emissions at or above these heights.

Conversely, in H2 the lower emission height means that the cloud has a much greater impact on the local area. The near-surface in Iceland is severely affected by both  $SO_2$  and  $SO_4$  in this scenario. The different winds at lower levels also result in very high levels of  $SO_4$  over Scandinavia in addition to the band across Germany and Italy seen in the other simulations. These factors explain the increase in the areas experiencing Very High concentrations in the maximum concentration graph in Figure 3. H2 has no cloud at FL300, as all of the emissions are emitted at lower altitudes and there is no significant uplift in this meteorological situation.

For the increases in emission duration in scenarios T1 and T2, the changes in the near-surface concentrations are not as large as seen in the M and H scenarios. This implies that the emissions are caught up in the same meteorological systems throughout this period. There are likely to be other cases with more dynamic/changing meteorology where changing the duration of emission would result in a much greater impact.

At FL300 the longer release durations lead to a more elongated plume downwind of lceland than the initial run, impacting a larger area of airspace with Very High concentrations. The maps in Annex D show that after the first 6 hours, the continuing T1 and T2 emissions interact with different winds to the initial emission. Consequently, although the early part of the plume follows the same path as the initial run, the tail of the plume exhibits different behaviour and a Very High concentration  $SO_2$  cloud moves south-eastwards over central Europe in the longest release scenario (T2). This transport and the resulting location of the cloud are controlled by the meteorology.

For the highest concentration thresholds, T1 and T2 result in a similar or reduced impact compared to the initial run. This is likely related to the longer duration releases, which result in the maximum concentrations being diminished. For the Moderate to High thresholds however the longer persistence of the cloud, due to the longer release period, contributes to a larger area being affected.

## 8.2 Method 2: Monte Carlo evaluation of uncertainty

This section presents an elementary exploration of the effects of stochastic variations in some key controlling factors that might influence peak volcanic gas concentrations over the UK during a future fissure eruption on Iceland. Here, probability distributions for various peak  $SO_2$  concentrations are derived from simple Monte Carlo simulations, anchored to the case study results produced by the detailed dispersion modelling in Section 6. The aim is to give a preliminary perspective on possible peak concentration levels that might be encountered under certain synoptic weather conditions, in the absence of more detailed modelling and analysis. A fundamental assumption relied on in here, is taken from the findings of the model sensitivity tests:

The maximum  $SO_2$  concentration experienced both near the surface and at altitude is linearly related to the amount of mass emitted in this particular meteorological situation. Further work is needed to understand whether this is the case in all meteorological situations and in more complex emission scenarios.

The findings of this analysis are expressed in terms of estimates of peak SO<sub>2</sub> concentrations and corresponding Air Quality Index (AQI) Band definitions.

## 8.2.1 Basics

Conventional variational calculation techniques are applied in a model using Monte Carlo re-sampling of uncertain volcanological and meteorological factors (represented in the model as RVDs – random variable distributions) to gauge their potential influence on the concentration of gas over the UK in a future prolonged fissure eruption from an Icelandic volcano. A schematic flowchart for the procedure, in Figure 8.6, summarizes this simplified conceptual model. In essence, it involves the convolving of volcanic source variations with variations due to wind transport to estimate plausible concentration distribution spreads over the UK.

For the purposes of this analysis the presumed controlling factors are: eruption mass and intensity (with exponentially-distributed variable Volcanic Explosivity Index, VEI, used as a surrogate parameter for both); eruption duration (the length of time the bulk of mass is ejected at source); and average overall north-westerly wind speed, which serves to indicate, crudely, the extent to which the gas column is 'bent over' at source and the resulting plume then remains concentrated or becomes laterally diffused, as it moves towards UK. Other volcanological and meteorological factors are held constant for the relevant Case Study scenario.

In the Monte Carlo model, UK gas concentration values are scaled relative to reference near-surface concentrations indicated by the NAME dispersion modelling (see Section 6.7 for details on the Case Studies, and Table 8.6 for related probability density functions PDFs). The Monte Carlo simulations involved 5,000 samples drawn with the Latin hypercube procedure from the input uncertainty distributions. The latter are based on parameterizations derived from the expert elicitation on effusive Iceland eruptions scenarios (Loughlin et al., 2013), or from plausible physical variations on meteorological factors (see Table 8.6).



Figure 8.6: Schematic model for estimating UK SO<sub>2</sub> concentration variations and exceedance probabilities, due to volcanic source and wind transport variabilities.

Random variable	PDF	Parameter(s)	Range/ truncation	Units
Eruption VEI	Expon.	0.54	$3 \rightarrow 4$	VEI
Climax duration	Gamma	K = 4.0; µ =2.0 {PDF*SQRT[VEI - 2.9]}	0.1 → 20	hours
Ref conc. 'Peak' site	Normal	μ = 1566; σ = 150	200 → 8000	μg/m <sup>3</sup>
Ref conc. N site	Normal	μ = 279; σ = 100	100 → 4000	μg/m <sup>3</sup>
Ref conc. E site	Normal	μ = 1070; σ = 120	200 → 6000	μg/m <sup>3</sup>
Ref conc. W site	Normal	μ = 396; σ = 120	100 → 6000	μg/m <sup>3</sup>
Ave. wind- speed	LogNorm	μ = 12; σ = 1.5	$5 \rightarrow 20$	m/s

Table 8.6: Principal random variables used in Monte Carlo simulations for estimating stochastic variability of peak SO<sub>2</sub> concentrations, determined from Loughlin et al (2013) and this work.

To enable uncertainty analysis on gas plume concentrations over the UK, three sites have been identified for extraction of time series data from the modelling work undertaken. Potential 'case study' episodes were chosen by inspecting daily maximum concentrations of each species, and a short-list was compiled of all times when these values were significantly elevated. For each episode on the short list, the hourly sequences were then examined in more detail in order to determine a limited number of case studies, representing significant episodes in terms of both magnitude of the concentration and duration of the episode.

The primary reference concentration value is the 'peak' maximum in a sequence, anywhere over the UK. To supplement this location peak value with information that gives some indication of spatial extent, geographically, three fixed locations were chosen, as shown on Figure 6.10; these are referred to as North, East and West Sites in the following discussion (see also Table 1 for definitions of parameter variation distributions).

#### 8.2.2 Dispersion Model Scenario

For the present variational analysis, the near-surface scenario Case Study 2 (Section 6.7.2) is selected, which is characterized as (AQI) 'Very High near-surface SO<sub>2</sub>'. This case study involves a period in which the UK experiences two episodes of elevated SO<sub>2</sub>, separated by around 12 days. The first episode lasts about two days and mainly involves hourly concentrations in the AQI Low Bands, although moderate levels are briefly reached in parts of Scotland. In the second episode, however, the maximum concentration reached in the UK region is 1566  $\mu$ g/m<sup>3</sup>. This is within the AQI Very High 10 Band, with hourly means showing that the bulk of the high concentration SO<sub>2</sub> plume is over Central and Eastern England.

The modelled air history maps suggest that the  $SO_2$  plume over the east of England was due to emissions from Iceland two days earlier. Notably, this represents a faster travel time than in most other modelled case studies. In the emission sequence for this period, there are only lower level emission layers, all within the troposphere. Moreover, total emissions on this day are relatively low compared to other days. But, in comparison with other eruption days, this day's emission amount is an order of magnitude higher than the mass of  $SO_2$  emitted in the same tropospheric height range on days with much more vigorous eruptions. In other words, it represents a weak plume in terms of altitude reach but carries similar quantities of gas to more buoyant plumes.

In this scenario, the consequent transport to the UK also occurs predominantly within the same tropospheric altitudes; scenario wind-speed information for these altitudes is given in Section 6.7.2.

## 8.2.3 Peak concentration distributions

N.B. in what follows, Monte Carlo model output values are given verbatim with a precision that is greater than is justified for the problem; they should be regarded as illustrative, and not definitive to the superficial precision.

For Case Study 2, simulation results for the 'peak' concentration location are shown in Figure 8.7, together with tabulated indicative distribution statistics. In this case the expected (mean) peak concentration is 2160  $\mu$ g/m<sup>3</sup>, somewhat greater than the reference value obtained from the dispersion modelling for this scenario (1566  $\mu$ g/m<sup>3</sup>). The corresponding modal (i.e. most likely) peak value is 2029  $\mu$ g/m<sup>3</sup>. Such concentration values correspond to AQI Band 11 'Hazardous'.

The main reason for these higher values in the distribution moments is that the source conditions for this case study scenario involved below average expected mass emission, based on elicited volcanic parameters for an effusive eruption of this type. Thus there is significant likelihood that a future event, with the same synoptic wind-field conditions, might produce a larger mass of gas emissions and higher peak concentrations; this simulation suggests there is about a 5% chance the peak concentration could exceed 3594  $\mu$ g/m<sup>3</sup>, or AQI Band 12 'Very hazardous'.



Stat	Conc µg/m^3
Minimum	955
Mean	2160
Maximum	5771
Std Dev	717
5th Perc.	1310
95th Perc.	3594
Mode	2029

Figure 8.7: Monte Carlo simulation distribution for Case Study 2: peak concentration, with tabulated statistics.

For the fixed East site location, the corresponding simulation distribution is shown on Figure 8.8. While the mean peak in this case, 1475  $\mu$ g/m<sup>3</sup>, is also elevated relative to the scenario reference (1070  $\mu$ g/m<sup>3</sup>), the mode 1188  $\mu$ g/m<sup>3</sup> is much closer. All three values correspond to AQI Band 10 'Very high'.



The 5% probability exceedance level is at about 2454  $\mu$ g/m<sup>3</sup>, AQI Band 11 'Hazardous'.

Stat	Conc. µg/m^3
Minimum	596
Mean	1475
Maximum	4820
Std Dev	494
Mode	1188
5th Perc.	888
95th Perc.	2454

Figure 8.8: Monte Carlo simulation distribution for Case Study 2: East site concentration, with tabulated statistics.

For the fixed West site location, the corresponding simulation distribution is shown on Figure 8.9. While the mean peak value in this case, 549  $\mu$ g/m<sup>3</sup>, is also elevated relative to the scenario reference (396  $\mu$ g/m<sup>3</sup>), the mode 408  $\mu$ g/m<sup>3</sup> is almost identical. The mean value corresponds to AQI Band 7 'High', whereas the mode is AQI Band 5 'Moderate'.



The 5% probability exceedance level is at about 1023  $\mu$ g/m<sup>3</sup>, AQI Band 9 'High'.

Figure 8.9: Monte Carlo simulation distribution for Case Study 2: West site concentration, with tabulated statistics.

1023

95th Perc.

For the fixed North site location, of the three the furthest away from the 'peak' location, the simulation distribution is shown on Figure 8.10. Again, the mean peak in this case, 396  $\mu$ g/m<sup>3</sup>, is elevated relative to the scenario reference (279  $\mu$ g/m<sup>3</sup>), and so is the mode, 375  $\mu$ g/m<sup>3</sup>. However, inspection of Figure 6 suggests the high mode in this case may be a slight artefact due to the limited number of Monte Carlo samples (5,000). These mean and mode values for the peak concentration correspond to AQI Band 5 'Moderate'.

In the tail of the distribution, the 5% probability exceedance level is at about 1023  $\mu$ g/m<sup>3</sup>, AQI Band 9 'High'.



Figure 8.10: Monte Carlo simulation distribution for Case Study 2: North site concentration, with tabulated statistics.

#### 8.2.4 AQI Probabilities

The preceding Monte Carlo simulation peak concentrations can be recast in terms of AQI Bands and the probabilities of different Bands being experienced in such synoptic conditions at the selected locations can be evaluated.

These probabilities are summarized, by Band and by locality, in Figures 8.11-8.14. Note that in this representation, the extreme tails of the concentration distributions can entail non-zero probabilities for higher Bands than those which correspond to  $95^{th}$  percentile values. Thus, for the 'peak' concentration scenario, there is about a 20% chance of occurrence of SO<sub>2</sub> concentrations in Band 12, 'Very hazardous' and for the East Site, about 3% chance of this happening.

On the basis of these simulations, the probabilities of high to hazardous concentrations at localities well away from where the peak concentration is found are very much lower or negligible (see Figures 8.13, 8.14).



Figure 8.11: Case Study 2: probabilities of AQI Bands in the peak concentration locality.



Figure 8.12: Case Study 2: probabilities of AQI Bands in the locality of East site.



Figure 8.13: Case Study 2: probabilities of AQI Bands in the locality of West site.



Figure 8.14: Case Study 2: probabilities of AQI Bands in the locality of North site.

#### 8.2.4.1 Country scale minimum AQI levels

Taking all four location simulations jointly, the above findings on AQI Band occurrence probabilities can be used to address the question: "what are indicative probabilities that an AQI Band will be exceeded at almost all localities on a country scale?". The answers are plotted in Figure 8.15 and values given in Table 8.7.



Figure 8.15: Case Study 2: probabilities for AQI Band exceedance at almost all localities on the country scale.

Band	Exceedance prob. "everywhere" In UK
AQI 3	29.7%
AQI 4	25.4%
AQI 5	19.9%
AQI 6	11.3%
AQI 7	9.8%
AQI 8	3.0%
AQI 9	0.8%
AQI 10	0.1%

 Table 8.7: Case Study 2: probabilities for AQI Band exceedance due to similar widespread peak concentration levels at almost all localities on UK scale

These findings suggest a probability of about 30% that Band 3 (or lower) will the common minimum, with reducing probabilities for higher Bands. By the same token, the probability of Band 10 'Very high' being experienced countrywide in an episode like this is estimated at about 0.1%, or 1-in-1000.

# 9 A basic assessment of the impact of the eruption on UK habitats using critical loads, critical levels and dynamic modelling

The air pollutants SO<sub>2</sub>, nitrogen oxides (NO<sub>x</sub>=NO+NO<sub>2</sub>) and ammonia (NH<sub>3</sub>) can contribute to acidification and adversely affect natural and semi-natural habitats. The sensitivity of habitats to acidification has been quantified in the UK (and across Europe) using "critical loads", defined as "a quantitative estimate of the exposure to one or more pollutants below which significant harmful effects on specified elements of the environment do not occur according to present knowledge" (Nilsson & Grennfelt, 1988). The amount of acid deposition that exceeds the critical load is called the "exceedance". Exceedance of the critical load is an indication that the ecosystem is at risk from potential harmful effects in the long term. Therefore, exceedance is not a quantitative estimate of "damage" to the environment; it does not necessarily mean that harmful or adverse effects have already occurred or may be observed, but that there is a risk of damage in the long-term. Critical loads are a concept for "long-term" protection of ecosystems; they do not provide information on the timescales for damage (when the critical load is exceeded) or "recovery" (when deposition is reduced below the critical load). Timescales for damage and recovery are affected by delays in changes in the chemical environment in soil and vegetation due to various buffering processes, and "biological" delays in the responses of organisms due to persistence, dispersal and establishment processes. The models to predict biological delays are not well developed, but chemical processes are better understood and have been captured in dynamic models. Under this project the "Very Simple Dynamic" model (VSD: Posch & Reinds, 2009) has been applied to illustrate the effects of volcanic sulphur deposition on an example habitat (dwarf shrub heath). Critical loads refer to the pollutant deposition load, usually expressed as a rate of influx per year.

At high concentrations, gaseous pollutants can also have direct impacts on habitats. A "critical level" of gaseous concentration above which damage has been observed has been established for many pollutants. Exceedance of the critical level for  $SO_2$  was also assessed in the study. Although the critical load concept is aimed at evaluating chronic pollutant deposition rather than increased deposition within a single year (or shorter timescale), it provides a useful context for assessing the relative importance of a volcanic S deposition episode.

The assessment of potential impacts on the environment was carried out using two scenarios:

- (i) baseline concentrations and deposition for 2005
- (ii) average concentrations and deposition for a Laki-type eruption lasting 5 weeks within the same year (2005)

Scenario (ii) utilises the EMEP4UK model run for 2005, which simulates eight eruptions in the year. The annual mean concentrations and deposition for one "average" eruption are calculated from the baseline plus one-eighth of the simulated annual additional deposition from the multiple eruptions. These values are referred to as the "average Laki-type scenario" in this section.

It should be noted that there are uncertainties in the estimates of emissions, concentrations, deposition, critical loads and habitat areas. Some uncertainties can be quantified, while others cannot (Skeffington et al, 2007); the results of this study do not take account of any uncertainties in the input data or models used.

## 9.1 Methods

#### 9.1.1 Critical loads and exceedances

Critical loads for acidification are calculated and mapped in the UK for nine habitat types (Hall et al, 2015a): acid grassland, calcareous grassland, dwarf shrub heath, montane, bog, managed coniferous woodland, managed broadleaved woodland, unmanaged woodland and freshwaters. Both managed and unmanaged woodlands are included since the long-term protection of the whole ecosystem function (i.e. soils, trees, linked aquatic ecosystems) is important. Managed woodland is assumed to be primarily productive forest where harvesting and removal of trees takes place, whereas "unmanaged woodland" is assumed to be "managed" only for biodiversity or amenity rather than timber production. Further details on the methods for calculating UK critical loads can be found in Hall et al. (2015a).

Maps of the terrestrial habitat distributions used for national critical loads research for Defra are based on a combination of the CEH Land Cover Map 2000 (Fuller et al, 2002(a)(b)) and additional data sets such as species distribution data, soils and altitude (Hall et al, 2015a). However, it should be noted that the habitat distribution maps and areas used for UK critical loads research (a) only include areas where data exist for the calculation or derivation of critical loads; (b) may differ from other national habitat distribution maps or estimates of habitat areas. Critical loads (and habitat areas) are mapped on a 1x1km grid of the UK, with the habitat areas in each grid square being derived from the Land Cover Map. For surface freshwaters, the areas included in the critical load exceedance analysis are based on the catchment areas of a total of 1752 sites only, predominantly in acid sensitive regions, across the UK (Hall et al, 2015a).

The calculation of acidity critical load exceedance takes into account the contributions from both sulphur and nitrogen (oxidised and reduced) deposition. Details on the methods for calculating exceedance can be found in Hall et al (2015a,b); the exceedance metrics used in this study are:

- The area of each habitat with exceedance of critical loads
- The percentage habitat area exceeded
- The Accumulated Exceedance (AE)
- The Average Accumulated Exceedance (AAE)

The percentage area of habitats with exceedance of critical loads is a useful metric for illustrating pollutant impacts, but responds little to small changes in deposition rate since the only areas affected are those with a current deposition rate that is around the critical load.

AE takes into account both the area exceeded and the magnitude of exceedance and is calculated as:

AE (keq year<sup>-1</sup>) = exceedance (keq ha<sup>-1</sup> year<sup>-1</sup>) \* exceeded habitat area (ha)

While this metric is useful for comparing different scenarios, as the results are expressed in keq year<sup>-1</sup> they tend to be very large numbers and not intuitive to understand. It should also be noted that the same AE can arise from a large exceedance and small exceeded area, or a small exceedance and large exceeded area.

AAE averages the AE across the entire habitat area and provides a more intuitive value for comparing exceedances for different scenarios, and gives an indication of

the difference in magnitude of exceedance, even if there is no change in the percentage area of habitat exceeded. AAE is calculated as:

AAE (keq ha<sup>-1</sup> year<sup>-1</sup>) = AE (keq year<sup>-1</sup>) / total habitat area (ha)

#### 9.1.2 Critical levels and exceedances

The critical levels for SO<sub>2</sub> were established in 1992 (Ashmore & Wilson, 1993; CLRTAP 2014). Values have been assigned for four vegetation types (Table 9.1); in this study the annual mean concentrations for cyanobacterial lichens, forest and semi-natural ecosystems have been used. The critical level for SO<sub>2</sub> used in the EU Air Quality Framework Directive Daughter Directives is an annual mean of 20  $\mu$ g/m<sup>3</sup> to protect ecosystems. The total UK land area exceeding the two different threshold values (10 and 20  $\mu$ g/m<sup>3</sup>) has been calculated for the baseline and the average Laki-type scenario. Although critical levels can be applied to individual habitats, for this impact assessment only the total land area of the UK exceeding the thresholds has been calculated; the areas exceeding critical levels are small (see Results) and therefore a more detailed analysis is unnecessary.

Vegetation type	Critical level SO <sub>2</sub> [μg/m <sup>3</sup> ]	Time period(s)
Cyanobacterial lichens	10	Annual mean
Forest ecosystems	20	Annual mean
(including understorey vegetation)		Half-year mean (Oct-Mar)
(Semi-) natural	20	Annual mean
		Half-year mean (Oct-Mar)
Agricultural crops	30	Annual mean
		Half-year mean (Oct-Mar)

Table 9.1: Critical levels for SO<sub>2</sub> ( $\mu$ g/m<sup>3</sup>) by vegetation category (CLRTAP, 2014).

#### 9.1.3 Dynamic modelling

Dynamic models typically require large amounts of site-specific input data. The VSD model (Posch & Reinds, 2009) has been developed in Europe to enable dynamic modelling to be carried out at regional or national scales using fewer input parameters. In the UK the VSD has previously been applied to a number of habitat types sensitive to acidification (Evans et al, 2012). In this study it has been applied to areas of dwarf shrub heath in the UK. The model uses critical load input parameters and additional data on soil characteristics and chemistry. It also requires a time series of historic and projected deposition data (e.g. from 1870 to 2100). In Evans et al (2012) the VSD used a time series of deposition generated using the FRAME (Dore et al, 2007; Matejko et al, 2009) model, with data calibrated to present day Concentration Based Estimated Deposition (CBED; ROTAP 2012). Deposition scenarios for the current study were derived from the EMEP4UK model (Simpson et al, 2012; Vieno et al, 2010). Historic time series of deposition were generated by rescaling the FRAME sequences to the EMEP4UK baseline data for 2005. For the average Laki-type scenario, the 2005 values in the time series were replaced by the scenario deposition. A limitation of the current VSD model application (MS Access) is that each model run is restricted to a maximum of nine specified years, one of which must be the final year of the time series (2100). Three model runs were needed to generate the model outputs at 10 yearly intervals from 1870 to 2100, plus the additional year of 2005 for the baseline and average Laki-type scenario.

The VSD outputs of relevance to this study are soil pH and ANC (Acid Neutralising Capacity). These parameters are calculated for each 1x1 km square of the habitat distribution for which model input data are available (92%), over a timeline of every

10 years over the period 1870 to 2100, plus 2005 for the baseline and average Lakitype scenario. Results are summarised by calculating percentiles of the outputs for all (67295) habitat squares.

## 9.2 Results

#### 9.2.1 Deposition and concentration data

The annual average concentration and deposition data were provided from the EMEP4UK model and consisted of:

- (a) baseline data for 2005
- (b) a scenario for an average Laki-type eruption lasting 5 weeks

The annual mean SO<sub>2</sub> concentrations across the UK land area are 2.02  $\mu$ g/m<sup>3</sup> for the baseline, and 2.23  $\mu$ g/m<sup>3</sup> for the average Laki-type scenario. Values range from 0.26 to 38.2  $\mu$ g/m<sup>3</sup> for the baseline and 0.35 to 38.5  $\mu$ g/m<sup>3</sup> for the eruption scenario. The highest concentrations are seen across England (Figure 9.1).



Figure 9.1: EMEP4UK modelled SO<sub>2</sub> concentrations for (a) baseline scenario; (b) average Laki-type scenario.

The deposition data for each scenario consist of two sets of values: (i) deposition to moorland or grassland (i.e. low-growing vegetation); (ii) deposition to woodland. These data are the sum of wet plus dry deposition and the values for different vegetation types reflect the different rates at which dry deposition occurs on different vegetation. Total acid deposition is used in the critical load exceedance and VSD calculations and is represented by the sum of both sulphur and nitrogen deposition; values for "moorland" are applied to the non-woodland habitats, and values for "woodland" to the woodland habitats. The average Laki-type scenario increases the amount of sulphur in deposition (Table 9.2).

	S deposition budget (kT S year <sup>-1</sup> )				
	Bas	eline	Average Laki-type		
	Moorland	Woodland	Moorland	Woodland	
England	65	50	80	64	
Wales	12	13	17	19	
Scotland	39	43	80	84	
NI	6	6	10	10	
UK	122	112	187	177	

NB. To convert from kT S year<sup>-1</sup> to keq ha<sup>-1</sup> year<sup>-1</sup> divide by 0.0016

Table 9.2: Non-marine sulphur deposition budgets (kT S year<sup>-1</sup>) assuming moorland and woodland across the entire UK land area, for the baseline and average Laki-type eruption scenario

The maps of the modelled S deposition to moorland vegetation (Figure 9.2) clearly show the increase in S deposition across the upland areas of the UK for the average Laki-type scenario, with the highest inputs from the volcanic eruptions depositing across Scotland.



Figure 9.2: EMEP4UK modelled sulphur deposition assuming moorland vegetation everywhere for (a) baseline scenario; (b) average Laki-type scenario.

#### 9.2.2 Exceedance of acidity critical loads

This section summarises the results of the ecosystem impact assessments; a full set of the results by country and habitat are summarised in the Annex E. It should be noted that the exceedance results presented in this report for the 2005 baseline scenario are based on EMEP4UK modelled deposition, and differ from exceedance results carried out under Defra contract AQ0826, which are based on the deposition calculated using the Concentration Based Estimated Deposition (CBED: RoTAP 2012). The critical loads data are identical in both studies. The baseline scenario shows 22.3% of the UK habitat area had exceedance of the acidity critical loads. For reference, the CBED annual average deposition for 2004-06, results in 58.3% of UK habitats having exceedance of the acidity critical loads (Hall et al, 2015b). These differences are due to differences in the deposition methodologies (Carslaw et al, 2013).

When the average Laki-type scenario is modelled, the exceeded area increases from 22.3% (baseline) to 51% (Table 9.3). In the baseline scenario, the area exceeded was lowest in Scotland, whereas for the Laki-type scenario the areas exceeded were highest in Scotland (Table 9.3, Figure 9.3). This shows that the areas predicted to have the highest deposition following an eruption are in Scotland. In terms of habitats, the largest difference was for dwarf shrub heath, with 15.6% of the area exceeded across the UK for the baseline scenario, increasing to 66% with the average Laki-type scenario (Table 9.4). This is because 82% of the dwarf shrub heath mapped in the UK occurs in Scotland. The largest increase in AAE was also predicted for Scotland, from 0.05 keq ha<sup>-1</sup> year<sup>-1</sup> for the baseline scenario, to 0.21 keq ha<sup>-1</sup> year<sup>-1</sup> for the average Laki-type scenario. The largest increases in AAE for the UK between the baseline and average Laki-type scenario were for the montane, dwarf shrub heath, bog and acid grassland habitats.

	Total area of sensitive habitats	Bas	eline	Average	Laki-type
	(km²)	%ex	AAE	%ex	AAE
England	18635	28.2	0.10	34.6	0.14
Wales	7798	37.8	0.13	52.6	0.22
Scotland	48083	17.6	0.05	57.9	0.21
NI	3537	19.8	0.05	39.8	0.11
UK	78051	22.3	0.07	51.0	0.19

Table 9.3: Summary of exceedance of acidity critical loads for sensitive habitats: percent habitat area exceeded (%ex) and AAE (keq ha<sup>-1</sup> year<sup>-1</sup>) by country for the baseline and average Laki-type scenario.

Habitat*	Area (km <sup>2</sup> )	Bas	eline	Average	Laki-type
		%ex	AAE	%ex	AAE
Acid grass	15336	41.9	0.13	70.7	0.28
Calc. grass	1808	0.0	0.0	0.0	0.0
Heath	24705	15.6	0.03	66.0	0.21
Bog	5454	27.9	0.08	74.5	0.24
Montane	3054	47.8	0.17	91.6	0.49
Conifer	8374	23.8	0.10	34.4	0.14
Broadleaf	7452	14.5	0.06	18.4	0.08
Other wood	4011	9.4	0.03	13.3	0.05
Freshwaters	7857**	8.6	0.03	12.5	0.05
All habitats	78051	22.3	0.07	51.0	0.19

\*Abbreviated names used in this column. Full names: acid grassland, calcareous grassland, dwarf shrub heath, managed coniferous woodland, managed broadleaved woodland, Unmanaged woodland.

\*\*based on the catchment areas of 1752 catchments only.

Table 9.4: Summary of exceedance of acidity critical loads: habitat area exceeded (%ex) and AAE (keq  $ha^{-1}$  year<sup>-1</sup>) by habitat for the UK for the baseline and average Laki-type scenario.



Figure 9.3: Acidity Average Accumulated Exceedance (AAE) for all habitats combined for (a) baseline; (b) average Laki-type scenario.

#### 9.2.3 Exceedance of critical levels of SO<sub>2</sub>

The UK land areas with annual average SO<sub>2</sub> concentrations that exceed the critical levels (10 and 20  $\mu$ g/m<sup>3</sup>) are the same for both the baseline and the average Laki-type scenario, and are very small (Table 9.5). The region with the largest area exceeded is England. The mean concentrations of SO<sub>2</sub> are 2.02  $\mu$ g/m<sup>3</sup> and 2.23  $\mu$ g/m<sup>3</sup> for the baseline and average Laki-type scenario respectively. Typical annual average concentrations in England in 2005 were in the range 2-6  $\mu$ g/m<sup>3</sup> (see AURN annual statistics on UK-Air for measurements).

	Land area	Critical lev	/el 10 μg/m³	Critical level 20 μg/m <sup>3</sup>	
	(km²)	Area	% exceeded	Area	% exceeded
		Exceeded		Exceeded	
England	131152	434	0.33	85	0.07
Wales	20761	0	0.0	0	0.0
Scotland	78744	20	0.03	0	0.0
NI	14177	25	0.18	0	0.0
UK	244834	479	0.20	85	0.04

Table 9.5: Exceedance of critical levels of  $SO_2$  by land area exceeded (km<sup>2</sup>) and % area exceeded; results are the same for the baseline and average Laki-type scenario.

#### 9.2.4 VSD model outputs for dwarf shrub heath

Figures 9.4 and 9.5 compare the VSD predictions for areas of UK dwarf shrub heath habitat under the baseline and average Laki-type scenario for soil pH and ANC. They show that the average eruption scenario could depress soil pH from 4.56 to 4.48 and depress ANC from 8.7 to -2.9 meq/m<sup>3</sup>. Such an acidity pulse tends to leach base cations from soil, but this effect was not predicted to greatly affect soil buffering capacity in the long term, as shown by the rapid recovery of pH and ANC to nearly

the baseline values by 2010. It should also be noted that these are 50<sup>th</sup> percentile values (Figures 9.4 and 9.5) and therefore do not represent the most sensitive sites, such as those on weakly buffered soils that have not yet recovered from 20<sup>th</sup> century sulphur pollution (Evans et al, 2014).



Figure 9.4: 50th percentile of modelled ANC (meq/m<sup>3</sup>) for areas of UK dwarf shrub heath habitat for the baseline scenario and average Laki-type scenario.



Figure 9.5: 50th percentile of modelled soil pH for areas of UK dwarf shrub heath habitat for the baseline scenario and average Laki-type scenario.

Maps of the modelled soil pH and ANC for 2005 (Figures 9.6 and 9.7) clearly show the spatial patterns and potential changes in values between the baseline scenario and the average Laki-type scenario. These changes in soil conditions may not appear to be severe, but many organisms reach their limits of acidity tolerance in the range from pH 4 to pH 4.5 and even short-term drops in pH can have drastic effects such as fish kills in streams (Evans et al, 2014). Recovery from any biological impacts could take longer than the chemical recovery of the pH and ANC to pre-eruption values.



Figure 9.6: Modelled pH in 2005 for areas of UK dwarf shrub heath for (a) the baseline scenario; (b) the average Laki-type scenario.



Figure 9.7: Modelled ANC in 2005 for areas of UK dwarf shrub heath for (a) the baseline scenario; (b) the average Laki-type scenario.

# 10 Halogen acids and H<sub>2</sub>S

Impacts of a volcano plume in the far field are a direct result of the chemical composition of the emitted volcanic gases and aerosol phase chemicals (particulate matter, PM). The plume composition changes with time as the volcano plume components interact both with the other volcano plume components (Martin et al., 2008; Martin et al., 2012; Pyle et al., 2009) and with the components of the background atmosphere as the plume is physically and chemically processed in the atmosphere with time (Allard et al., 2005; Fischer et al., 1998). HCl, HF and H<sub>2</sub>S are known volcanic plume constituents as are many other trace species (Table 10.1). These three species are of particular interest for differing reasons: HCl will add to the acidity of the plume, HF has known corrosive and biological effects and H<sub>2</sub>S is a toxic pollutant. All three are co-emitted with SO<sub>2</sub> to varying degrees, but little information is available regarding the possible levels which could occur in the far field and hence there is much uncertainty concerning them.

Species	Chemical symbol
Water	H <sub>2</sub> O
Carbon dioxide	CO <sub>2</sub>
Sulphur dioxide	SO <sub>2</sub>
Hydrogen sulphide	H <sub>2</sub> S
Hydrogen fluoride	HF
Hydrogen chloride	HCI
Hydrogen bromide	HBr
Hydrogen iodide	HI
Bromine oxide	BrO
Mercury	Hg (elemental and speciated)
Nitric acid	HNO <sub>3</sub>
Nitrogen dioxides	NO <sub>2</sub>
Ammonia	NH <sub>3</sub>
Carbonyl sulphide	COS
Helium	Не
Nitrogen	N <sub>2</sub>
Argon	Ar
Metals including:	In, Rb, Cs, Be, B, Cr, Ni, Cu, Mo, Cd, W, Re,
	Ge, As, In, Sn, Sb, Te, Tl, Pb,
	Mg, Sr, Sc, Ti, V, Mn, Fe, Co, Y, Zr, Hf, Ta, Al,
	P, Ga, Th, U, La, Ce, Pr, Nd, Sm,
	Eu, Gd, Tb, Dy, Er, Tm

Table 10.1: Volcanic plume constituents. See Oppenheimer et al. (2003) for further details.

## *10.1 Literature summary*

This short report assesses the method by which initial levels of the halogen acids and  $H_2S$  can be estimated and identifies initial levels to be used in this short modelling project. It is noted that the literature review carried out was non-exhaustive as a full literature survey was beyond the scope of this project.

Volcanoes emit many different chemical species of gases. To understand the impact that volcanic gases will have on the environment, it is important to know the proportions (ratios) of different gas species, and the total amount which is emitted ("flux"). Fluxes of some gases can be measured more easily than others, in particular  $SO_2$ . Often it is only possible to measure the flux of  $SO_2$ , while the fluxes of other gases are calculated or inferred based on their proportional abundance relative to  $SO_2$ . Techniques to measure gas concentrations or ratios of the chemical concentration with respect to a plume tracer (in this case  $SO_2$ ) are summarised in

Table 10.2. Overall more than fifty literature records were found for measurements of the chemical species of interest and these are summarised in Annex F. It is noted that due to time constraints, primarily references were sourced from the review by Aiuppa (2009) and references therein, and that other reviews of the literature exist e.g. Pyle et al. (2009) published in the same volume as Aiuppa (2009). In most of these studies the ratios of HCI, HF and H<sub>2</sub>S to SO<sub>2</sub> are reported rather than the absolute concentration, hence this is the value using molar quantities which is summarised in this report. Where it was not reported as this ratio, the ratio was calculated.

Abbreviation	Methodology	Direct/offline/ indirect sampling	Off-line/ <i>In-situ</i> analysis
FP	Filter Pack	Offline	Off-line
FTIR	Fourier transform infrared spectroscopy	Direct	In-situ
DT	Diffusion Tubes	Off line	Off-line
UVS	UltraViolet Spectroscopy, including COSPEC and DOAS	Direct	In-situ
L	Leachate of fresh ash	Indirect	Off-line
GS	Gas sensors, e.g. electrochemical or mass spectrometry	Direct	In-situ

Table 10.2:Summary of measurement techniques for halogen acids and hydrogen sulphide in volcano plumes

In the ideal situation, chemical concentrations would be measured directly in the gas phase in the plume as it emerges from the volcano and forms an atmospheric plume. There are obvious logistical reasons why in many cases this is not possible. Studies which assess or measure the halogen acid concentrations in volcano plumes close to the eruption location can be separated into several distinct method types: Direct methods in which the measurements of the gases are made in-situ either by spectroscopy, electrochemical sensors, off-line methods where sampling is taken in the field and subsequently subject to laboratory analysis. Indirect methods where a proxy for the volcano gas composition is used, typically either a leachate of the ash fallen from the plume or gas bubbles trapped in solidified magma. Studies which used gas sampled from bubbles in solidified magma are not included in this summary due to probable disconnection from atmospheric processing in the near field plume. for example sulphur oxidation during cooling and mixing as a opposed to the sulphur staying in a reduced form is not a well constrained process. It noted that these studies do have important information regarding the magma chemistry before it is erupted. In addition satellite/remote sensing studies were not included in this literature review mostly due to the coarse resolution of the information at, or near, volcanic sources and also the limited scope of this report.

Direct measurements have been made of gas compositions using Fourier transform infrared spectroscopy (FTIR), gas sensors (e.g. electrochemical sensors). Ultraviolet spectroscopy (UVS) techniques are usually the most direct however may not be made very close to the initially emitted plume due to access issues. This also applies to sampling for offline analysis (e.g. diffusion tubes (DT) or filter packs (FP) or

bubblers.). Ash is collected either during or after an eruption and the water soluble chemicals washed off the ash, into a leachate solution. The composition of the leachate is then analysed in the laboratory and the composition used as a proxy for the composition of gases erupted in the plume.

#### 10.1.1 HCI:SO<sub>2</sub> and HF:SO<sub>2</sub> ratios

There is wide range of ratios of  $HCI:SO_2$  in the literature, as can be seen in Annex F, from 0.01 to 12.5. This range covering several orders of magnitude is due to geological and atmospheric reasons including variations in the halogen content of the magma from which the gas was emitted, the mixing of the emitted volatiles with the background atmosphere as it is emitted and cools, location and residence time in atmosphere of sampling and to some extent method of sampling. Measurements cover a wide range of techniques, volcanic settings and time resolutions therefore an approach to narrow the range to look at the range of values which are applicable to modelling a Laki-type eruption has been made.

Even within a single eruption sequence the volatile ratios, HCI:SO<sub>2</sub> and HF:SO<sub>2</sub> are likely to be highly variable. However in order to initialise the EMEP4UK model runs a reasonable estimate was attempted. In order to rapidly assess which ratios would be appropriate for initialising the volcano modelling for the Icelandic Laki-type eruption the following criteria were applied:

- I. Magma type must be similar to Iceland, i.e. basaltic or basaltic andesitic
- II. Direct, off-line or indirect, or fresh leachate analysis methodology.

Using these criteria the number of literature measurements were reduced to 11, summarised in Table 10.3. The selected studies come with some caveats: the studies cover a range of tectonic settings, not all of which are directly analogous to the Icelandic geological setting, e.g. some are subduction zone volcanoes, not all are reported in the peer review scientific literature (e.g. the Burton et al. data reported in Bagnato et al. (2013)) and some methods are not ideal for studying plume composition, e.g. Bagnato et al. (2013), which is stated by the authors themselves. However this approach was used as a practical rationalisation to see the range of values that can occur given that the aim of this modelling was to do a first assessment. It is again noted that other off-line analyses have been made using scoria and aerosols, e.g. Ilyinskaya et al. (2012).

The range of HCI:SO<sub>2</sub> values are 0.03-1.69 and HF:SO<sub>2</sub> 0.03-0.34. The arithmetic and geometric mean molar ratios for HCI:SO<sub>2</sub> were  $0.47\pm0.42$  and 0.31 respectively and for HF:SO<sub>2</sub> 0.09±0.1 (1SD) and 0.05 respectively. Though these statistics are not meaningful for a particular volcano or modelling a specific event, they are useful as indicators. The composition of a volcano plume can change within an eruption process and interannually, for example Aiuppa et al. (2007) showed this clearly for Etna eruptions over several years and this was also the case in the 2010 eruption of Eyjafjallajokull.

As this modelling exercise is focussed on an Icelandic eruption, the studies based around the 2010 eruption are looked at in a little more detail. Bagnato et al. (2013) collected fresh and aged ash and performed leachate measurements during the 2010 eruption of Eyjafjallajokull and commented on the evolution of the chemical implications for the plume. The Bagnato et al. (2013) sampling points were downwind from the Eyjafjallajokull eruption (up to 50 km or ~1 hour with an assumed wind speed of 13 m s<sup>-1</sup>). The S:CI ratios are approximately constant and a possible decrease in the S:F ratio is reported (the subject of discussion in e.g. Goff et al.

		Magma				
Volcano	Country	composition	Method	HCI/SO2	HF/SO2	Reference
Yasur	Vanuatu	Basalt Andesite	FP	0.27	0.10	Allard et al. in Aiuppa (2009)
Kudryavy	Russia	Basalt Andesite	DS	0.28	0.02	Fischer et al (1998) in Aiuppa (2009)
Galunggung	Indonesia	Basalt/basaltic andesite	L	0.23	0.03	De Hoog et al. (2001) in Aiuppa (2009)
Poas	Costa Rica	Basalt to Dacite	DS	0.53	0.06	Symonds et al. (1994) in Aiuppa (2009)
Villarica	Chile	Basaltic Andesite	FTIR	0.33		Sawyer et al. (2011)
Villarica	Chile	Basaltic Andesite	FP	0.38	0.10	Aiuppa (2009) using Mather et al. (2004); Shinohara & Witter (2005); Witter et al (2004)
Villarica	Chile	Basaltic Andesite	FP	0.59	0.23	Mather et al. (2004)
Villarica	Chile	Basaltic Andesite	FP	0.70		Sawyer et al. (2011)
Eyjafjallajokull	Iceland	Basaltic	FTIR	0.25	0.01	Burton in Bagnato et al. (2013)
Eyjafjallajokull	Iceland	Andesitic (Allard)	FTIR	0.83	0.02	Allard in Bagnato et al. (2013)
Eyjafjallajokull	Iceland	Basaltic/basaltic andesitic	L	1.69	0.03	Bagnato et al. (2013)
Erta Ale	Ethiopia	Basaltic	FP	0.38	0.34	Allard et al. (2004) in Aiuppa (2009)
Hekla	Iceland	Basaltic icelandite	SS	0.06	0.09	Hunton et al. (2005) and Moune et al. (2007) in Aiuppa (2009)
Kilauea	Hawaii	Basaltic	FP, DS	0.03	0.03	Aiuppa (2009) using Crowe et al. (1987); Gerlach (1993); Greenland et al. (1985); Miller et al. (1990); Olmez et al. (1986)

(2001) and Greenland et al. (1985)). The decrease in S:F is apparently non-linear close to source (Figure 10.1 and Figure 10.2).

Table 10.3 Studies used in assessment of HCI:SO<sub>2</sub> ratio appropriate for Icelandic volcano modelling scenario.



Figure 10.1: Elemental ratios of S, Cl and F in leachate downwind from Eyjafjallajokull eruption (figure from Bagnato et al. (2013)).



Figure 10.2: Literature summary of S/CI ratios for the Ejyafjallajokull eruption (figure from Bagnato et al. (2013)).

The ratio of HCI:SO<sub>2</sub> and HF:SO<sub>2</sub> in the gas phase will be highest at the plume emission point, and back-extrapolation to the initial value will have high uncertainty level. Bagnato et al. (2013) state that the leachate analyses in their study (or in others) are not representative of the gas phase plume composition and Eyjafjallajokull summit magma is very different from Eyjafjallajokull flank eruption magma. It is noted that the paper is recently the subject of some discussion (Goff et al., 2001; Greenland et al., 1985). The FTIR measurements (cited and discussed in Bagnato et al. (2013) but unpublished) would be more applicable and should be investigated further if possible. The only direct gas measurements of SO<sub>2</sub> and HCI gas were taken during the Hekla 2000 eruption (Rose et al., 2006) with aircraft instruments in an aged plume. Although not directly comparable with the 2010 eruption, purely for illustrative purposes the S:CI ratios for the two sets of measurement are summarised in Figure 10.3 in which it can be inferred that the aged plume is significantly depleted in chloride.



Figure 10.3: S:CI Molar ratio as a function of time (Bagnato et al. (2013) data assumed wind speed 13 m.s<sup>-1</sup>).

## 10.1.2 H<sub>2</sub>S

There are significantly fewer studies and hence there is less evidence to work with for assessing which ratio of  $H_2S:SO_2$  should be used. Annex F summarises the literature and includes reference found within reports and reviews by Halmer et al. (2002), Shinohara (2013) and Aiuppa (2009). The range of ratios covers 0.01 - 2.86, with the only Icelandic measurement from Surtsey, Iceland with alkali-basalt reporting a  $H_2S:SO_2$  ratio of 0.04, Halmer et al. (2002). The values may vary over orders of magnitude lower or higher, therefore assessment of modelling results will need to take this into consideration. It would be useful to identify and collate any unpublished work from Icelandic eruptions for future studies.

#### 10.1.3 HBr

HBr emissions are locally important emissions in the troposphere and have been the subject of several recent studies (Aiuppa et al., 2009; Bobrowski et al., 2003; Bobrowski et al., 2007; Oppenheimer et al., 2006; Roberts et al., 2009; von Glasow et al, 2009), which followed the discovery by Bobrowski et al. (2003) of high BrO concentrations in volcano plumes and associated ozone depletion. However these studies have illustrated the complex nature of the bromine chemistry, initiated by the photolysis of HBr or the heterogeneous uptake of HBr onto atmospheric aerosol: volcanic ash or sulphate aerosol in the context of a volcano plume. Without implementing the full bromine chemistry in the EMEP4UK model one could not realistically follow the bromide emitted chemically downwind in the plume. Figure 10.4 from Roberts et al. (2009), shows the loss mechanisms for HBr, but it is also noted that there are HBr formation mechanisms via reaction of atomic bromine with gas phase formaldehyde or the HO<sub>2</sub> radical. Therefore due to the short and scoping nature of this project, it is proposed not model Br separately.



Figure 10.4: Schematic of the autocatalytic BrO formation cycle in volcanic plumes showing BrO formation, ozone destruction and HNO3 formation. If the plume becomes depleted in HBr, there is a switch in the aerosol-phase reactions to produce BrCl instead of Br<sub>2</sub> (Roberts et al., 2009).

#### 10.2 Ratios for modelling

This brief literature review has shown that there is a limited literature for direct measurements of halogen gases and  $H_2S$  close to the emission point volcanoes for good operational reasons. This work does not take into account the modelling literature and several other sources of information and in the future better parameterisations may be available through these and other routes.

The ratios to be used in the EMEP4UK model runs are summarised in Table 10.4. For the EMEP4UK modelling of the potential impacts of an Icelandic effusive eruption, an upper conservative estimate was selected. Specifically  $HCI:SO_2 = 1$ , and  $HF:SO_2 = 0.3$  (for  $HCI:SO_2$  this is approximately the arithmetic mean plus one standard deviation). The  $HCI/SO_2$  is at the high end of the range summarised in Table 10.3. It is noted that there are few measurements and also that the fluoride content of the Eyjafjallajokull eruption was highly variable, so results from the modelling will need to be interpreted with this in mind.

For the EMEP4UK modelling of the potential impacts of an Icelandic effusive eruption, an upper conservative estimate was selected. Specifically  $H_2S:SO_2 = 0.04$ .

It is strongly emphasised that these are estimations of a conservative nature, bearing in mind the aim of the modelling being the impact of a future eruption on the UK. No attempt is made here to look at the consequences of the range of concentrations presented or their application to impact assessment.

Chemical Ratio	Selected value
HCI:SO <sub>2</sub>	1
HF:SO <sub>2</sub>	0.3
$H_2S:SO_2$	0.04

Table 10.4: Chemical ratios selected.

#### 10.3 Thresholds for HCI, HF and H<sub>2</sub>S

Table 10.5 summarises air quality levels for the halogen gases and  $H_2S$  as provided by IVHN and Defra (see links below Table 10.5).

Compound	Averaging period	Level
Hydrogen fluoride (HF)	1 hour	0.2 ppm/ 0.16 mg/m <sup>3</sup> (1)
Hydrogen chloride (HCI)	1 hour	0.5 ppm/0.75 mg/m <sup>3</sup> (1)
Hydrogen bromide (HBr)	1 hour	0.2 ppm/ 0.7 mg/m <sup>3</sup> (1)
Hydrogen sulphide $H_2S$	24 hour	150 μg/m <sup>3</sup> (2)

(1) http://www.ivhhn.org/index.php?option=com\_content&view=article&id=83;

(2) http://archive.defra.gov.uk/environment/quality/air/airquality/publications/halogens/fullreport.pdf

Table 10.5: Summary of Defra and IVHN published human health air quality values for the halogen acids and hydrogen sulphide.

#### 10.4 Set up of EMEP4UK for modelling HCI, HF and H<sub>2</sub>S

The current EMEP4UK chemical scheme does not parameterise the full HCl chemistry (previous EMEP4UK versions used EQSAM scheme which did have this) and it also does not have HF or  $H_2S$  explicitly in it.

For the purposes of this study a soluble species,  $X_{sol}$  and two insoluble unreactive species,  $X_{insol}$  were modelled.  $X_{sol}$  was assigned the cloud scavenging, wet deposition and dry deposition values used for nitric acid (HNO<sub>3</sub>).  $X_{sol}$ :SO<sub>2</sub> was 1:1 ratio, as per the HCI:SO<sub>2</sub> suggested from Section 10.2. Table 10.6 summarises the Henry's Law solubility (k<sub>H</sub>) properties of the relevant volcanic species. Note there is very limited literature on HF therefore for this work HF is also approximated by the  $X_{sol}$  value.

 $X_{insol}$  high and low are conservative tracers in the model with no reactions and no deposition. However if the  $X_{insol}$  tracer molecule leaves the modelling domain, the chemical is removed. Therefore concentration of  $X_{insol}$  will reflect the dispersion of the plume. This simplified parameterisation was due to time constraints in the modelling project however there is a lot of information in the modelled data can be derived from this modelled variable. The  $X_{sol}$  and  $X_{insol, high}$  provide a range of possible values for HF as the datasets provide possible lower and upper bounds for chemical species going from very soluble to insoluble. HF is a soluble gas therefore a first cut reasonable approximation is that the  $X_{sol}$  modelled values would be a better approximation to that which would be expected for HF in the atmosphere. The  $X_{insol,low}$  gives information for a lower concentration non-reactive species, e.g. H<sub>2</sub>S. H<sub>2</sub>S has a significantly longer atmospheric lifetime that SO<sub>2</sub>, though there is only a limited literature on this chemical in the atmosphere.

	k <sub>н</sub> [M/atm]	−d In k <sub>H</sub> /d(1/T) [K]	References
HNO <sub>3</sub>	2.4×10 <sup>6</sup> /K <sub>A</sub> *	8700	Brimblecombe and Clegg [1988]
HCI	2.0×10 <sup>6</sup> /K <sub>A</sub> *	9000	Brimblecombe and Clegg [1988]
SO <sub>2</sub>	1.2	3100	Pandis and Seinfeld [1989]
HF	9.6/K <sub>A</sub>	7400	Brimblecombe and Clegg [1988]
H <sub>2</sub> S	0.1	2000	Lide and Frederikse [1995]

\*For strong acids, the solubility is often expressed as kH = ([H+] + [A-])/p(HA). To obtain the physical solubility of HA, the value has to be divided by the acidity constant KA;

\*\* see Compilation of Henry's Law Constants for Inorganic and Organic Species of Potential Importance in Environmental Chemistry, R.Sander http://www.mpch-mainz.mpg.de/~sander/res/henry.htmland references therein. Note K<sub>A</sub> for HNO<sub>3</sub> and HCI are both large as they are strong acids.

Table 10.6: Summary of Henry's Law solubility constants for relevant gases.

# 10.5 Results

#### 10.5.1 X<sub>sol</sub> (proxy for HCl and HF): Hourly and daily statistics

Figure 10.5 summarises the hourly average, 95<sup>th</sup> percentile and maximum concentrations over the UK for the 10 year period for  $X_{sol}$ , the HCI and HF proxy (note: high upper limit for HF, probably approximately a factor of three lower). For the daily average concentrations are in general less than 1 µg/m<sup>3</sup> with highest concentrations being observed in the southern half of the UK and much lower, <0.5 µg/m<sup>3</sup> over much of Scotland. The 95<sup>th</sup> percentile values are also low, and in general less than 5 µg/m<sup>3</sup>. Maximum concentrations observed over the 80 eruptions are significantly higher and have a much more structured pattern indicating that these occurrences are caused by specific events rather than most of the eruptions, as discussed in previous parts of this report.

Highest concentrations can be observed in Scotland and southern England in the range of  $280 - 320 \ \mu g/m^3$ . For HCl these values are significantly below the human health limit of hourly exposure to  $750 \ \mu g/m^3$ . The values are slightly above the human health exposure limit of  $180 \ \mu g/m^3$ , however it is noted that the emission ratio of  $X_{sol}$ :SO<sub>2</sub> = 1 is approximately a factor of 3 higher than the ratio suggested in Section 10.1, therefore it is very unlikely that the HF human health exposure would be reached in any of the 80 scenarios modelled.



Figure 10.5: Hourly average  $X_{sol}$  results: Mean (upper panel); 95th percentile (middle panel) and maximum (lower panel); units:  $\mu g/m^3$ .

# 10.5.2 X<sub>insol.low</sub> and X<sub>insol,low</sub> proxy for H<sub>2</sub>S: Hourly and daily statistics

Figure 10.6 shows the mean, 95<sup>th</sup> and maximum hourly values for X<sub>insol</sub>, with the X<sub>insol</sub>:SO<sub>2</sub> ratio of 0.04 and 0.3 values (LHS and RHS respectively). These results are for a non-reactive tracer, which approximately covers the range of ratios for H<sub>2</sub>S. Average concentrations are <0.28 and <1.2  $\mu$ g/m<sup>3</sup> across the UK for X<sub>insol, low</sub> and X<sub>insol,high</sub> respectively. Maximum values are <35 and <130  $\mu$ g/m<sup>3</sup>. Daily average concentrations for specific sites are discussed below, but it is noted that the results from this work imply there is a very low risk of significant H<sub>2</sub>S concentrations over the UK following eruption, particularly if the emissions are at the low end of the X<sub>insol</sub>:SO<sub>2</sub> range. Even at the high end of the range likely concentrations are not a risk to human health, using the metrics in Table 4. However as with the halogen acids, other effects have not been considered in this work.



Figure 10.6: Hourly average Xinsol results.LHS: Xinsol:SO<sub>2</sub> = 0.04, RHS Xinsol:SO<sub>2</sub> = 0.3; Mean (upper panel); 95th percentile (middle panel) and maximum (lower panel); units:  $\mu$ g/m<sup>3</sup>.

#### 10.5.3 Site specific case studies

Table 10.7 summarises the statistics for the daily average  $X_{sol}$  concentrations over the 10 year dataset for thirty sites across the UK ( $\mu$ g/m<sup>3</sup>) (Figure 10.7). These sites from the Defra UK national network (UK Eutrophying and Acidifying Atmospheric Pollutants Acid Gas and Aerosol Network) have been selected specifically so that the modelling output can be compared against ambient concentrations (Tang et al, 2013). All daily average  $X_{sol}$  concentrations are below 10  $\mu$ g/m<sup>3</sup>. For both HF and HCI these would be very low concentrations. Annex G provides equivalent tables for  $X_{insol}$ .

The data for 6 example sites for 4 years (2003-2006) have been produced (Annex H). An example is shown in Figure 8.10 (note that a log scale is used on panels B-E in each figure). In panel B  $X_{sol}$  is shown as HCl and compared against the ambient average concentration derived from measurements. In these figures the eruption is show for reference in the top panel, and the sulphur species SO<sub>2</sub> and sulphate for both the baseline model run and the volcano scenarios. In the bottom panel, an attempt to relate the HCl to the emitted HCl has been done by ratio, according to the equation below, which has the baseline SO<sub>2</sub> subtracted from the volcano model scenario. It is noted that this is only an indicative as there are both model variations in both runs which mean sometimes the volcano run results in less SO<sub>2</sub> than the baseline (results >0 not shown) and also very high HCl:SO<sub>2</sub> occur when the difference between the SO<sub>2,volcano</sub> and SO<sub>2, base</sub> are very similar, which sometimes are anomalies.

$$\frac{HCl}{SO_2} = \frac{HCl}{SO_{2,volcano} - SO_{2,base}}$$

Three coastal sites were assessed (Shetland, Goonhilly and Stoke Ferry) and for most of the eruptive periods the daily average concentration is lower than monthly ambient average concentration, indicating that the volcano plume would not be causing a noticeable change in concentrations. During eruptive periods there are occasions when significantly higher daily average HCl concentrations are seen, for example in Figure 10.8 it can be seen at the end of August 2003 there are three days when there are significantly elevated HCI concentrations. Note this particular event is discussed in earlier parts of this report in one of the Case studies. Inland sites such as Auchencorth and Harwell have lower background concentrations of HCI whereas. London has a slightly higher HCI ambient concentration than the rural background sites. It can be seen in all cases and sites, during eruptions when the plume reaches the UK surface there are significant increases in HCI concentrations. These higher concentrations are not hazardous with respect to human health, using the published metrics summarised in Table 10.5 but no assessment of these concentrations against other effects or metrics has been considered here but it is recommended that this should be perhaps an area for further research. Generally the HCI:SO<sub>2</sub> is significantly <1, with some ratios between 0.1 -1 but mostly <0.1. The volcano plume is present at the UK surface after the eruption stops in some cases and it can be seen that there is some mixing of volcanic plume within the background air much of the time, even when the plume is not directly having a high impact, leading to slightly elevated levels of sulphur species much of the time. This would be an interesting area to further explore these modelling results.



Figure 10.7: UK Eutrophying and Acidifying Atmospheric Pollutants Acid Gas and Aerosol Network (UKEAP AGANET) site locations.
	mean	SD	Median	Мах	P90	P95	P99
Rosemaund	0.45	2.33	0.01	58.4	0.81	1.93	7.06
Narberth	0.38	1.8	0.01	35.08	0.7	1.59	6.43
Halladale	0.23	0.92	0.01	22.55	0.47	1.05	4.2
Auchencorth	0.25	1.11	0.01	29.62	0.47	1.02	3.93
Shetland	0.27	1.19	0.01	23.12	0.49	1.11	5.23
Glensaugh	0.26	0.97	0.01	19.07	0.55	1.19	4.47
Moor House	0.29	1.3	0.01	45.81	0.57	1.26	5.13
Rothamstead	0.43	1.99	0.02	54.44	0.88	1.76	6.82
Strathvaich Dam	0.25	1.1	0.01	26.67	0.49	1.11	4.85
Eskdalemuir	0.26	1.09	0.01	29.3	0.48	1.11	4.85
High Muffles	0.3	1.16	0.02	19.17	0.62	1.36	5.66
Stoke Ferry	0.45	2.02	0.02	52	0.9	1.91	8.05
Yarner Wood	0.37	1.75	0.01	50.66	0.7	1.57	5.99
Cromwell Road	0.35	1.47	0.02	35.04	0.73	1.49	5.29
Sutton	0.43	2.08	0.01	49.44	0.83	1.8	7.04
Bonington Lagganlia	0 27	1 22	0.01	34 03	0.52	1 23	4 82
Hillsborough	0.28	1 21	0.01	33 24	0.56	1.23	4 71
Lough Navar	0.28	1 22	0.01	33.92	0.54	1 19	4 9
Rum	0.26	1.1	0.01	27.63	0.53	1.14	4.86
Edinburgh	0.25	1.11	0.01	29.62	0.47	1.02	3.93
Cwmystwyth	0.38	1.85	0.01	45.62	0.68	1.62	6.24
Carradale	0.22	0.96	0.01	21.48	0.44	0.93	4
Barcombe_Mills	0.33	1.07	0.02	16.22	0.74	1.63	4.95
Detling	0.36	1.52	0.02	34.08	0.75	1.61	5.68
Harwell	0.48	2.37	0.02	64.21	0.94	2.02	7.32
Ladybower	0.36	1.56	0.01	30.68	0.73	1.67	6.22
Plas_Y_Brenin	0.33	1.59	0.01	42.85	0.6	1.35	5.88
Caenby	0.41	1.66	0.02	33.33	0.82	1.8	7.2
Goonhilly	0.36	1.47	0.02	37.01	0.75	1.52	6.42

Table 10.7: Summary statistics for the daily average  $X_{sol}$  concentrations over the 10 year dataset for thirty sites across the UK (µg/m<sup>3</sup>).



Figure 10.8: EMEP4UK 24 hour average results summary for Cromwell Road, London 2003: A: Volcano emissions (Tg); B: Modelled (purple squares) and measured average ambient (orange line) HCl; C: Modelled SO<sub>2</sub>: Baseline (continuous), eruption scenario (crosses); D: Modelled SO<sub>4</sub>: Baseline (continuous), eruption scenario (crosses); E: Ratio of HCl to SO<sub>2</sub>: eruption scenario emission ratio (purple line); eruption scenario-baseline (green crosses).

## **10.5.4** More detailed chemistry and further work

There is a significant amount of further information which can be derived from this dataset and it is noted that comparison of the surface composition of EMEP4UK can be compared against the baseline model run to understand the addition loading of the volcano emissions on the baseline PM levels rather than just the sulphate. An example of this is show for one short episode at 2 sites (Marylebone Road, London and Auchencorth Moss, Scotland) in Figure 10.9. The chemical composition of the gas phase and aerosol phase can be seen to change due to the eruption on the 16-18 August, but for the London site, the PM levels are similar to baseline levels a few days before. The chemical titration of the gas phase due to the stronger acidity of the sulphate. There is probably significant further work which can be done on the chemical composition of aerosol when the UK is impacted by volcano emissions as it is likely that the pH of the aerosol is highly dependent on the age of the plume.



Figure 10.9: Comparison of EMEP4UK model output for August 2003: in each plot the first bar is from the base run, the second from the volcano eruption model run. Top graphs show gas phase SO<sub>2</sub> (red), ammonia (orange) and NO<sub>x</sub> (green) and the bottom panel show particulate phase species (see legend for details). The LHS graphs are for London Marylebone Road and the RHS for Auchencorth Moss, Scotland.

# 11 Conclusions

The aim of this work was to generate and assess the hazards from a "precautionary worst case scenario" for an Icelandic effusive eruption, which could then be used for impact assessment, decision making and preparation in advance of an eruption. To achieve this, information from the literature and the findings of an expert elicitation have been synthesised to determine appropriate eruption source term parameters and associated uncertainties. One scenario has then been used to create a limited ensemble of model simulations of the dispersion and chemical conversion of the emissions of volcanic gases during such an eruption. This has utilised the high resolution modelling of the Met Office's NAME Lagrangian dispersion model and the Centre for Ecology and Hydrology's EMEP4UK Eulerian model. Modelling outputs have addressed the likelihood of near-surface concentrations of sulphur and halogen species exceeding specified health thresholds; concentrations at aviation relevant altitudes have also been evaluated. In addition, the effects of acid deposition of volcanic species on ecosystems have been assessed.

The differences between the model outputs in both Section 6 and 7 provide one indication of the uncertainty in these results. In addition, the findings of Section 8 suggest that an order of magnitude uncertainty is feasible due to possible variations in the eruption source term. Such a factor of ten uncertainty is the difference between the Moderate4 threshold and the Very Hazardous12 threshold for both  $SO_2$  and  $SO_4$  on the Air Quality Index scale. This means that higher concentrations and longer duration pollution episodes cannot be excluded from consideration.

Summaries of each of the strands of work, together with conclusions and implications are given below.

### Eruption Source Term

Eruption source terms for sulphur dioxide emissions for a 'Laki-type' eruption have been produced using a Monte Carlo analysis, with relevant source term factors estimated using the findings of an expert elicitation. Outputs comprise databases of multiple stochastic model runs, each holding sequences of 24-hour source terms for a Laki-type eruption, extending over a period of 6 weeks. These include information on plume height and SO<sub>2</sub> mass erupted for a height profile of five layers for each time-period, which allows a more realistic emission variation with height. These multiple scenarios could be used in other analysis work for the H55 risk or other investigations related to Laki-type eruptions. One scenario, representing a "precautionary worst case", has been chosen for use in the modelling. Although lower than the Thordarson and Self (2003) emission values, this source term better accounts for uncertainties in the emissions by pooling expert opinions.

A "model source term" has then been produced from this eruption source term by assuming that each spot height in the eruption source term is representative of a vertical layer. The five spot heights allow the definition of five vertical layers when assumptions are also made about the heights of zero emission. For lower altitude emissions, the five layers have been reduced to three. The definition of layers is essential to allow the representation of a realistic vertical eruption profile, which is an assumption in the emission derivation. The resulting vertical profiles can be used by any atmospheric chemistry and transport model, not just those used in this project.

The model source term comprises five weeks of daily emissions followed by one week of no emissions. Each day in the 5-week eruption has a different vertical profile and a different emitted mass of  $SO_2$ . The maximum  $SO_2$  emission on any one day is 2.248 Mt and the maximum height of emission on any day is 16.4 km above sea level

(assuming that the volcanic vent is 600 m above sea level). Assuming that any model source term layer wholly above 10 km asl is in the stratosphere then ~24% of SO<sub>2</sub> is emitted into the stratosphere and ~76% is emitted into the troposphere.

The model source term produced here reflects the outputs from the expert elicitation and better accounts for uncertainties in the emissions. It should be noted that an "eruption source term" defined with a different number of spot heights would result in a different "model source term", which could also have an influence on the results of the modelling.

#### Modelling

A limited study has been conducted with two atmospheric chemistry and transport models. This approach enables some of the uncertainty and the variability present due to the meteorology, chemistry and transport model, and source of driving NWP data to be addressed. In order to sample different meteorology, which naturally varies on seasonal, annual and decadal timescales, the 6-week model source term has been repeated eighty times over 10 years of meteorology. No two 6-week periods overlap. No uncertainty in the eruption emissions profile has been considered.

Both of the models used have limitations in their set-ups and certain assumptions have been made to produce these simulations. As such it is important not to consider that one model is right and the other wrong. No attempt has been made to combine the results into one ensemble; rather the results are presented separately to give an indication of the uncertainty.

Many of the tables in this report give the maximum values seen anywhere in the UK and Republic of Ireland region. These are not necessarily representative of the whole region and the maps should be referred to for more detail. A major limitation with looking at the maximum values in this way is that they can be strongly dominated by outliers. This is one reason why the NAME and EMEP4UK values in the tables are different. Analysis of the maps in the Annexes demonstrates that actually the geographical distribution and magnitudes of values are generally very similar between the two models.

A variety of results have been produced for  $SO_2$  and  $SO_4$  and are expressed relative to the UK Air Quality Index scale. All particulate sulphate is discussed without reference to its neutralisation state, but near the surface it is likely to exist as ammonium sulphate and in aged plumes the particles will be much less acidic than freshly formed sulphuric acid particles.

Health impacts from  $SO_2$  are linked to short-term acute exposures, which is why the UK AQI for  $SO_2$  is based on a 15-minute mean concentration. However, at the distance of the UK from Iceland (1000+ km) any plume will be reasonably well mixed by the time it reaches the surface, so such short term fluctuations are unlikely and the hourly concentrations presented here will be representative of the 15-minute averages that would be observed at monitoring sites and compared to the  $SO_2$  index.

#### Near Surface Results

The results show that for the majority of time (at least 95%) during the effusive eruption simulations near-surface concentrations of  $SO_2$  and  $SO_4$  over the UK are within the Low air quality index bands for both hourly and daily averages. However, on occasions, concentrations within the Moderate, High and Very High bands are possible for  $SO_2$  and concentrations up to and including Very Hazardous are possible for  $SO_4$ . For  $SO_2$  the number of simulations where the Moderate4 level is reached at any time is small (3 simulations out of 160 across both models for daily data and 38

out of 160 for hourly data) and the maximum consecutive duration of each episode is relatively short (approximately 1 day based on hourly data). For SO<sub>4</sub>, the occurrence of Moderate4 threshold exceedances is more frequent in the simulations (56 simulations out of 160 across both models for daily data and 120 out of 160 for hourly data) with a longer maximum consecutive duration of up to 9 days (based on daily data). These relative frequencies and durations decrease as the thresholds increase.

In general during any 6-week eruption period only a relatively short time in total is spent above the AQ index Moderate4 threshold. In combination with the maximum duration data, this demonstrates that the UK is unlikely to be affected by week after week of significantly elevated concentrations; rather there will a number of short (hours to days) pollution episodes where concentrations could be elevated above Moderate4. This pattern fits with the generally changeable nature of the weather in the UK. Total and consecutive durations are longer for  $SO_4$  than  $SO_2$ , and can be particularly lengthy in the Low air quality index bands (1-2 weeks), which may be of relevance to health impact assessments.

The occurrence of above-Moderate episodes is related to both the weather pattern at the time and the emissions day that the plume is related to. A conducive weather pattern on a day with a low mass emission rate will not necessarily lead to a Moderate or above episode. The case studies demonstrate that pollution episodes can occur in a range of weather patterns and at different lead-times from the responsible volcanic emissions.

The fastest modelled time after the start of an eruption that a gas and aerosol plume arrives at the surface on the north-western borders of the UK is ~29 hours. For the south-east the arrival time is longer at about 2.5 days. There is a large spread of arrival times after the start of eruption, which is due to the meteorology during each of the simulation periods. It is possible that a plume from an individual day of emissions could reach the UK faster.

The maximum concentrations produced by the two models show that NAME simulates higher maximum  $SO_4$  than EMEP4UK over a wide area. The reason for this difference is unclear, particularly as the maximum  $SO_2$  concentrations are more similar, but there are a number of differences in model set-up that could contribute including:

- The horizontal and vertical distribution of the emissions at the source
- Differences in chemical conversion rates between the models
- Different treatment of oxidant species in the two models
- Difference in wet deposition parameterisations

The maximum maps from both models clearly show individual plume features, suggesting that these maps should be treated as being representative of the sorts of concentrations that could occur, rather than providing any specific details on geographical distribution.

Differences between the two models in the case-studies are also not surprising, but the agreement in plume transport shows that the two meteorologies used by the models are similar. It should be noted that the three case studies were chosen based on times when substantially elevated concentrations were seen in the NAME results, case studies based on similar events in the EMEP4UK data would also be expected to show differences between the models.

#### Flight Altitude Results

Similar to the near surface results the flight altitude results show that for the majority of time (at least 95%) during the effusive eruption simulations concentrations of  $SO_2$  and  $SO_4$  at all flight levels over the UK are within the Low air quality index bands for hourly averages. However the results also show that in all simulations across both models  $SO_2$  entered into the Moderate air quality banding at some point during the eruption at all cruising and on-route flight levels over the UK (FL240, FL300, FL370). Taking these two results together shows that in general exceedance durations must be short when Moderate4 and above levels are reached. The case studies help demonstrate that plumes are indeed very transient and do not remain over one location for long. Their location is entirely dependent on the meteorology, which at upper flight altitudes can be strongly controlled by the position of the jet stream. First arrival times of a plume over the UK can be as little as 6-7 hours from the start of eruption at upper flight levels if the jet stream direction is from lceland to the UK. This is much quicker than arrival times near the surface.

At higher altitudes the chance of exceeding higher  $SO_2$  thresholds increases, because most of the gas is emitted from the volcano at these heights. For  $SO_4$ greater concentrations are more likely at lower altitudes. The duration of exceedance has a similar trend with longer durations possible at upper altitudes for  $SO_2$  and at lower altitudes for  $SO_4$ . This is in part related to the time required for the conversion of  $SO_2$  to  $SO_4$ . In rapidly moving upper level plumes much of the  $SO_2$  will still be present when the plume reaches the UK. As a consequence the maximum consecutive duration of exceedance of the High threshold is up to 95 hours for  $SO_2$ , with all of the UK showing the potential to experience 20-40 hours at High  $SO_2$  air quality levels at cruise and on-route altitudes. This is considerably longer that the durations seen at the near-surface.

Analysis of the maps in the Annexes demonstrates that the geographical distribution and magnitudes of values are very similar between the two models. For example, there is a clear north-south gradient over the domain (and the UK) shown by both models for both the relative frequency and duration data. Areas closer to Iceland are more likely to experience higher concentrations of SO<sub>2</sub> and longer episode durations. The highest values are seen to the east of Iceland in both models which corresponds to the dominant wind flow regime in this region. This pattern matches previous assessments of meteorology and the hazard from volcanic ash eruptions on Iceland (e.g. Leadbetter and Hort, 2011).

As with the near surface results, the maximum concentrations produced by the two models show that NAME simulates higher maximum  $SO_4$  than EMEP4UK over a wide area at most flight levels. The reason for this difference is unclear, particularly as the maximum  $SO_2$  concentrations are fairly similar at all levels, but the same differences in model set-up mentioned above could be contributing.

The maximum maps from both models also clearly show individual plume features, suggesting that these maps should be treated as being representative of the sorts of concentrations that could occur, rather than providing any specific details on geographical distribution. In particular where "fingers" of plumes can be seen it may be more appropriate to draw a contour around them and consider this wider area as having the potential to experience that concentration (e.g. Fig 11.1).



Figure 11.1: The white line provides a better representation of the region where the black level concentrations could be experienced and avoids the risk of over-interpreting the significance of concentration "fingers" caused by individual plumes.

The difference in the layers used to represent the flight levels in the outputs from the two models may also have an influence. It is noticeable in the relative frequency maps in Annex C that in the lowest two levels for the EMEP4UK data the SO<sub>4</sub> data appears to be dominated by the anthropogenic signal. Only at FL150 does a volcanic signal really become apparent. In the NAME data, the lowest two levels show a clear topographic pattern. This is because, in the dynamic definition of flight layers based on pressure levels that is used in NAME, FL010 and FL070 can be below the ground surface in regions of high topography. These findings suggest that it may be more appropriate to use the near-surface results for these lower flight layers.

Case study 2 shows that a complex four-dimensional picture can occur with elevated concentrations occurring in different flight levels over different locations at different times. This has the potential to prove challenging for airspace management if the levels modelled here are considered to be of concern in further impact assessments.

#### Uncertainties

Many factors influence the ability to accurately forecast possible near-surface peak SO<sub>2</sub> concentrations over the UK in a future effusive Icelandic eruption. These include lack of knowledge about the eruption emissions (mass of each species, height of emission, and variability of these with time); the evolution of these species over time due to complexities in the atmospheric chemistry; the weather situation that would be present and how accurately this would be reflected by numerical weather prediction models. Whilst the bulk of the work in this report has addressed a "precautionary worst-case" scenario, the results should not be considered to represent the only possible outcome due to the inherent uncertainties involved.

Two, limited, assessments of some of these uncertainties have been conducted. One using a dispersion modelling approach, the other a simplified stochastic variational approach. Each has been conducted for one (different) meteorological case study period only. These reveal that plausible changes in some of the factors lead to significant variation in the simulated concentrations downwind. The times and order of arrival of the  $SO_2$  and  $SO_4$  plumes over the UK/Europe are also affected. The uncertainty assessments show that the factors having a first order impact on concentrations at the surface over the UK during a real eruption are the height of emission, the mass of the emission and the meteorological situation.

For example, the modelling assessment shows that the maximum near-surface concentration is strongly dependent on the height of the emission. Emissions primarily into the stratosphere, as in the H1 scenario, are mainly kept aloft (gases do not sediment out of the atmosphere unlike ash) and therefore have a small impact on the surface. Knowing the height of the emissions with respect to the tropopause will be a key input during a real eruption. In addition, the maximum SO<sub>2</sub> concentration experienced both near the surface and at altitude is linearly related to the amount of mass emitted in this particular meteorological situation. The modelling assessment demonstrates that maximum near-surface concentrations can vary by approximately an order of magnitude (i.e. a factor of 10) for SO<sub>2</sub> from just a factor of 2 change in emission heights and mass. For SO<sub>4</sub> the range is much greater, about a factor of 30.

Summing up the stochastic assessment findings for the selected case study, the 90% credible interval for a plausible peak near-surface  $SO_2$  concentration, somewhere over the UK, is 1300 - 3600 µg/m<sup>3</sup>. This range implies localized effects associated with AQI Band 10 'Very high' to Band 12 'Very hazardous'. However, on the balance of the probabilities enumerated here (i.e. at the median level), peak concentrations are unlikely to exceed Band 10. Note: The actual values generated are just indicative, perhaps order-of-magnitude estimates at best, and should not be invested with undue accuracy or precision – other factors could modulate these numbers, up or down.

It should also be noted that changes in the basic emission parameters can affect the geographical area experiencing a certain maximum AQI threshold. This can make a sizeable difference to the number of people who may be affected when considered on the European scale. For the UK, the stochastic approach suggests that there is a small, but non-zero probability (estimated at about 1%) that quite large areas of the country might experience peak SO<sub>2</sub> concentrations corresponding to AQI Band 9 or higher. At the median balance of probabilities, however, the countrywide peak concentration level is unlikely to exceed AQI Band 3 for the case study period.

The stochastic simulation outcomes, exploring broad variations in volcanic source output and in transport wind-speed, give an appropriate feel for the range of plausible peak concentrations that might be experienced and for their comparative probabilities of occurrence. It is believed that similar indicative findings would be obtained in terms of relative concentration levels and patterns for other gas species. These findings are for a short period (1-2 days) in the full eruptive source term scenario that involves quite modest amounts of SO<sub>2</sub> being injected into the lower atmosphere over Iceland. Quantifying concentration exceedance probabilities and uncertainty ranges for other eruption source conditions and other meteorological circumstances requires further work. This is the case for both of the methodologies considered here.

On the basis of the stochastic results for the single case study with a given form of eruption and one specific weather pattern, it is concluded that even an unspectacular fissure eruption on Iceland, expelling gas emissions into low altitudes for long enough, could be potentially problematic for the UK in terms of short duration peak concentrations.

#### Impact on UK habitats

The potential effects of a Laki-type eruption on exceedance of critical loads and critical levels, and on the dynamic effects over time (using VSD), have been assessed. The results compare annual average concentrations and deposition modelled in the EMEP4UK surface modelling with a selected baseline year, 2005. The total sulphur deposition to the UK land area for the average Laki-type scenario was approximately 55% greater than the baseline deposition in 2005. Sulphur deposition to moorland for the average scenario is 53% larger than the baseline (58% for the woodland deposition). While the increase in deposition may be linear, this represents a linear increase in pressure. The increase in impact is unlikely to be linear (see results for individual habitats in Table 9.4).

The largest annual average SO<sub>2</sub> concentrations over the UK occur across England; this is true for both the baseline scenario and the average Laki-type scenario. The land areas with concentrations above the critical levels are the same for both the baseline and the average Laki-type scenario, with 0.2% (479 km<sup>2</sup>) of the land area above 10  $\mu$ g/m<sup>3</sup> and 0.04% (85 km<sup>2</sup>) above 20  $\mu$ g/m<sup>3</sup>. These are very small areas of the UK total of 244834 km<sup>2</sup>, however they may be indicative that though the national effect may be small by area, there could potentially be significant effects at a local scale where these thresholds are exceeded.

The area of acid sensitive habitats with exceedance of acidity critical loads increased from 22.3% for the baseline scenario, to 51% for the average Laki-type scenario. Of the UK component countries, although Scotland had the lowest area of habitats with critical load exceedance under the baseline scenario, it had the highest area with exceedance for the average Laki-type scenario, reflecting the spatial pattern of the increased sulphur deposition from the eruption scenario. Dwarf shrub heath was the habitat in the UK with the largest increase in the area exceeded under the Laki-type scenarios; this is primarily due to 82% of the dwarf shrub heath habitat mapped in the UK occurring in Scotland.

The AAE for the UK was 1.7 times greater than the baseline for the average Lakitype scenario. The largest increase regionally was for Scotland, where the AAE increased from 0.05 keq ha<sup>-1</sup> year<sup>-1</sup> for the baseline scenario, to 0.21 keq ha<sup>-1</sup> year<sup>-1</sup> for the average Laki-type scenario. The largest increases in AAE across the UK between the baseline and average Laki-type scenario were for the montane, dwarf shrub heath, bog and acid grassland habitats.

The VSD model predicted that the increased sulphur deposition with the average Laki-type scenario caused a decrease in  $50^{\text{th}}$  percentile soil pH values in the year of the eruption from 4.56 to 4.48. The  $50^{\text{th}}$  percentile ANC values were similarly reduced, from 8.7 meq/m<sup>3</sup> to -2.94 meq/m<sup>3</sup>. As noted above, this is the  $50^{\text{th}}$  percentile and greater effects would be seen in sensitive areas. The implications of the punctuated acidity change in this scenario could be significant for freshwater natural systems and aquaculture.

This modelling work has demonstrated that a Laki-type eruption is likely to increase sulphur deposition in some parts of the UK, particularly in Scotland and some other upland regions. The increase in sulphur deposition has the potential to enhance the adverse impacts of acidification in these regions. However the limited nature of this assessment is noted and further investigation recommended.

#### Hazards from halogen gases and hydrogen sulphide

The EMEP4UK model was run over 10 years of meteorology with hydrogen chloride (HCI) modelled using HNO<sub>3</sub> dry and wet deposition parameters, and hydrogen fluoride (HF) and hydrogen sulphide (H<sub>2</sub>S) as non-reactive "upper" limit tracers. HCl and HF emissions of the same magnitude as SO<sub>2</sub>, modelled as a soluble species, result in average concentrations at the UK surface mostly <1  $\mu$ g/m<sup>3</sup> with a 95<sup>th</sup> percentile <5  $\mu$ g/m<sup>3</sup>. These average modelled HCl concentrations are similar to every-day coastal HCl concentrations. However there is large variability between the different eruption periods and maximum X<sub>sol</sub> concentrations over the UK can reach a few hundred  $\mu$ g/m<sup>3</sup>. The hourly human health limits for HF and HCl of 500  $\mu$ g/m<sup>3</sup> and 750  $\mu$ g/m<sup>3</sup> respectively are not exceeded in any of the simulations. HF is not as soluble as HCl, therefore the X<sub>insol, high</sub> species is useful as another proxy (emitted at a ratio of 0.3 rather than 1 to SO<sub>2</sub>). In this case the concentrations do not exceed ~30  $\mu$ g/m<sup>3</sup>.

 $H_2S$  modelled as an insoluble species emitted in the ratio range of 0.04 - 0.3 to  $SO_2$  also does not exceed the relevant human health 24-hour limit of 150 µg/m<sup>3</sup>. Daily average values using the high X<sub>insol</sub> ratio limit are generally <2 µg m<sup>-3</sup> with maximum modelled values of ~30 µg/m<sup>3</sup>.

The emission scenarios considered are insufficient to capture the entire range of variability in the numerous aspects of such a Laki-type eruption and in-particular only one set of ratios has been simulated so the values given should be taken as guidance only.

# 12 Acknowledgments

The Project Team acknowledge the help and assistance of representatives from Public Health England, Department of Health, Department for Transport and Department for Environment, Food and Rural Affairs in the definition of the parameters to be analysed in this work and the relevant thresholds to be used.

The Project Team would like to thank the members of the H55 Scientific Expert Advisory Group for their input and feedback on the report draft and for useful discussions at the meeting where these results were first presented.

# **13 Further Information**

This report is an amalgamation of twelve separate work package reports that were submitted by the Project Team members to the H55 Project Board during and at the end of the work in July 2014. For further information on specific aspects of the work contained here please contact the following.

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## 14 References

- Aiuppa, A., 2009. Degassing of halogens from basaltic volcanism: Insights from volcanic gas observations. Chemical Geology, 263, 99-109.
- Aiuppa, A., Baker, D.R., and Webster, J.D., 2009. Halogens in volcanic systems. Chemical Geology, 263, 1-18.
- Aiuppa, A. et al., 2007. The tropospheric processing of acidic gases and hydrogen sulphide in volcanic gas plumes as inferred from field and model investigations. Atmospheric Chemistry and Physics, 7, 1441-1450.
- Allard, P. et al., 2004. Magma supply rate to erta ale lava lake (afar) inferred from measured volatile and heat fluxes. Geophysical Research Abstracts, 6, 06601, SRef-ID: 1607-7962/gra/EGU04-A-06601.
- Allard, P., Burton, M., and Mure, F., 2005. Spectroscopic evidence for a lava fountain driven by previously accumulated magmatic gas. Nature, 433, 407-410.
- Ashmore, M.R. & Wilson, R.B. (eds), 1993. Critical levels of air pollutants for Europe. Background papers prepared for the ECE Workshop on critical levels, Egham, UK, 23-26 March 1992.
- Bagnato, E. et al., 2013. Scavenging of sulphur, halogens and trace metals by volcanic ash: The 2010 Eyjafjallajokull eruption. Geochimica Et Cosmochimica Acta, 103, 138-160.
- Bobrowski, N. et al., 2007. Reactive halogen chemistry in volcanic plumes. Journal of Geophysical Research-Atmospheres, 112.
- Bobrowski, N., Honninger, G., Galle, B., and Platt, U., 2003. Detection of bromine monoxide in a volcanic plume. Nature, 423, 273-276.
- Brimblecombe, P., and Clegg, S.L, 1988. The solubility and behaviour of acid gases in the marine aerosol. Journal of Atmospheric Chemistry, 7, 1-18.
- Carslaw, D., Chemel, C., Davis, L., Dore, T., Griffiths, S., Hall, J., Metcalfe, S., Redington, A., Sokhi, R., Sutton, P., Vieno, M., and Whyatt, D. 2013. Defra Phase 2 deposition model evaluation. Contract report to Defra. Kings College London.
- Chenet, A-L., Fluteau, F., and Courtillot, V., 2005. Modelling massive sulphate aerosol pollution following the large 1783 Laki basaltic eruption. Earth and Planetary Science Letters, 236, 721-731.
- CLRTAP, 2014. Manual on methodologies and criteria for Modelling and Mapping Critical Loads & Levels and Air Pollution Effects, Risks and Trends. http://wge-cce.org/Pubications/Mapping\_Manual
- Collins, W.J., Stevenson, D.S., Johnson, C.E., and Derwent, R.G., 1997. Tropospheric ozone in a global scale three dimensional Lagrangian model and its response to NOx emission controls, Journal of Atmospheric Chemistry, 26, 223–274, doi:10.1023/A:1005836531979.
- Crowe, B.M., Finnegan, D.L., Zoller, W.H., and Boynton, W.V., 1987. Trace-element geochemistry of volcanic gases and particles from 1983-1984 eruptive episodes of Kilauea volcano. Journal of Geophysical Research-Solid Earth and Planets, 92, 13708-13714.
- de Hoog, J.C.M., Koetsier, G.W., Bronto, S., Sriwana, T., and van Bergen, M.J., 2001. Sulfur and chlorine degassing from primitive arc magmas: temporal changes during the 1982-1983 eruptions of Galunggung (West Java, Indonesia). Journal of Volcanology and Geothermal Research, 108, 55-83.
- Dore, A.J., Vieno, M., Tang, Y.S., Dragosits, U., Dosio, A., Weston, K.J., and Sutton, M.A., 2007. Modelling the atmospheric transport and deposition of sulphur and nitrogen over the United Kingdom and assessment of the influence of SO<sub>2</sub> emissions from international shipping. Atmospheric Environment, 41, 2355-2367.
- ENTEC: Defra, 2010. UK ship emissions inventory, final report, http://ukair.defra.gov.uk/reports/cat15/1012131459\_21897\_Final\_Report\_291110.pdf.

- Evans, C., Hall, J., Rowe, E., Tipping, E., Henrys, P., Smart, S., Moldan, F.,
  Blomgren, H., Oulehle, F., Norris, D., Helliwell, R., Cosby, J., and Jenkins, A.,
  2012. Critical Loads and Dynamic Modelling Umbrella. Final Report to Defra
  under contract no. AQ0801. Centre for Ecology and Hydrology.
- Evans, C.D., Chadwick, T., Norris, D., Rowe, E.C., Heaton, T., Brown, P., and Battarbee, R.W., 2014. Persistent surface water acidification in an organic soil-dominated upland region subject to high atmospheric deposition: The North York Moors, UK. Ecological Indicators, 37, 304-316.
- Fischer, T.P., Giggenbach, W.F., Sano, Y., and Williams, S.N., 1998. Fluxes and sources of volatiles discharged from Kudryavy, a subduction zone volcano, Kurile Islands. Earth and Planetary Science Letters, 160, 81-96.
- Fuller, R.M., Smith, G.M., Sanderson, J.M., Hill, R.A., and Thomson, A.G. 2002a. The UK Land Cover Map 2000: construction of a parcel based vector map from satellite images. Cartographic Journal, 39, 115-25.
- Fuller, R.M., Smith, G.M., Sanderson, J.M., Hill, R.A., Thomson, A.G., Cox, R., Brown, N.J., and Gerard, F. 2002b. Countryside Survey 2000 Module 7: Land Cover Map 2000. Final Report, CSLCM Final CEH report to Defra.
- Gerlach, T.M., 1993. Oxygen buffering of Kilauea volcanic gases and the oxygen fugacity of Kilauea basalt. Geochimica Et Cosmochimica Acta, 57, 795-814.
- Greenland, L.P., Rose, W.I., and Stokes, J.B., 1985. An estimate of gas emissions and magmatic gas content from Kilauea volcano. Geochimica Et Cosmochimica Acta, 49, 125-129.
- Goff, F. et al., 2001. Passive infrared remote sensing evidence for large, intermittent CO2 emissions at Popocatepetl volcano, Mexico. Chemical Geology, 177, 133-156.
- Hall, J., Curtis, C., Dore, T., and Smith, R., 2015a. Methods for the calculation of critical loads and their exceedances in the UK. Unpublished report to Defra under contract AQ0826.
- Hall, J., and Smith, R. 2015b. Trends in critical load exceedances in the UK. Unpublished report to Defra under contract AQ0826.
- Halmer, M.M., Schmincke, H.U., and Graf, H.F., 2002. The annual volcanic gas input into the atmosphere, in particular into the stratosphere: a global data set for the past 100 years. Journal of Volcanology and Geothermal Research, 115, 511-528.
- Heard, I.P.C., Manning, A.J., Haywood, J.M., Witham, C., Redington, A., Jones, A., Clarisse, L., and Bourassa, A., 2012. A comparison of atmospheric dispersion model predictions with observations of SO<sub>2</sub> and sulphate aerosol from volcanic eruptions, Journal of Geophysical Research, doi:10.1029/2011JD016791, 117, D00U22.
- Highwood, E. J., and Stevenson, D. S., 2003. Atmospheric impact of the 1783–1784 Laki Eruption: Part II. Climatic effect of sulphate aerosol. Atmospheric Chemistry and Physics, 3, 1177–1189.
- Hunton, D.E. et al., 2005. In-situ aircraft observations of the 2000 Mt. Hekla volcanic cloud: Composition and chemical evolution in the Arctic lower stratosphere. Journal of Volcanology and Geothermal Research, 145, 23-34.
- Ilyinskaya, E., Martin, R.S., and Oppenheimer, C., 2012. Aerosol formation in basaltic lava fountaining: Eyjafjallajokull volcano, Iceland. Journal of Geophysical Research-Atmospheres, 117.
- Jones, A.R., Thomson, D.J., Hort, M. and Devenish, B., 2007. The U.K. Met Office's next-generation atmospheric dispersion model, NAME III, in Borrego C. and Norman A.-L. (Eds) Air Pollution Modeling and its Application XVII (Proceedings of the 27th NATO/CCMS International Technical Meeting on Air Pollution Modelling and its Application), Springer, pp. 580-589.

- Leadbetter, S., and Hort, M., 2011, Volcanic ash hazard climatology for an eruption of Hekla Volcano, Iceland, Journal of Volcanology and Geothermal Research, 199, 230-241.
- Lide, D.R. and Frederikse, H. P. R., 1995. CRC Handbook of Chemistry and Physics, 76th Edition. CRC Press, Inc., Boca Raton, FL.
- Loughlin, S.C., Aspinall, W.P.A, Vye-Brown, C., Hort, M., and Witham, C., 2013. Large magnitude fissure eruptions in Iceland: source characterisation. Report prepared for the Civil Contingencies Secretariat. British Geological Survey Open File Report OR/12/098.
- http://www.bgs.ac.uk/research/volcanoes/LakiEruptionScenarioPlanning.html Martin, R.S. et al., 2008. Composition-resolved size distributions of volcanic aerosols
- in the Mt. Etna plumes. Journal of Geophysical Research-Atmospheres, 113. Martin, R.S., Wheeler, J.C., Ilyinskaya, E., Braban, C.F., and Oppenheimer, C., 2012. The uptake of halogen (HF, HCI, HBr and HI) and nitric (HNO3) acids into acidic sulphate particles in quiescent volcanic plumes. Chemical Geology, 296, 19-25.
- Matejko, M., Dore, A.J., Dore, C.J., Blas, M., Kryza, M. Smith, R., and Fowler, D., 2009. The influence of long term trends in pollutant emissions on deposition of sulphur and nitrogen and exceedance of critical loads in the United Kingdom. Environmental Science and Policy, 12, 882-896.
- Mather, T.A., Tsanev, V.I., Pyle, D.M., McGonigle, A.J.S., Oppenheimer, C., and Allen, A.G., 2004. Characterization and evolution of tropospheric plumes from Lascar and Villarrica volcanoes, Chile. Journal of Geophysical Research-Atmospheres, 109.
- Mather, T.A., Witt, M.L.I., Pyle, D.M., Quayle, B.M., Aiuppa, A., Bagnato, E., Martin, R.S., Sims, K.W.W., Edmonds, M., Sutton, A.J., and Ilyinskaya, E., 2012. Halogens and trace metal emissions from the ongoing 2008 summit eruption of Kilauea volcano, Hawaii. Geochimica et Cosmochimica Acta, 83, 292-323.
- Miller, T.L., Zoller, W.H., Crowe, B.M., and Finnegan, D.L., 1990. Variations in tracemetal and halogen ratios in magmatic gases through an eruption cycle of the Puu-Oo vent, Kilauea, Hawaii – July-August 1985. Journal of Geophysical Research-Solid Earth and Planets, 95, 12607-12615.
- Moune, S., Sigmarsson, O., Thordarson, T., and Gauthier, P.-J., 2007. Recent volatile evolution in the magmatic system of Hekla volcano, Iceland. Earth and Planetary Science Letters, 255, 373-389.
- Nilsson, J., and Grennfelt, P., 1988. Critical loads for sulphur and nitrogen. Report 1988:15. UNECE/Nordic Council of Ministers, Copenhagen, Denmark.
- Olmez, I., Finnegan, D.L., and Zoller, W.H., 1986. Iridium emissions from Kilauea volcano. Journal of Geophysical Research-Solid Earth and Planets, 91, 653-663.
- Oman, L., Robock, A., Stenchikov, G. L., Thordarson, T., Kich, D., Shindell, D. T., and Gao, C., 2006. Modelling the distribution of the volcanic aerosol cloud from the 1783-1784 Laki eruption. Journal of Geophysical Research, 111, D12209, doi:10.1029/2005JD006899
- Oppenheimer, C., Pyle, D.M., and Barclay, J. (editors), 2003, Volcanic Degassing, Geological Society, London, Special Publication, 213.
- Oppenheimer, C. et al., 2006. BrO formation in volcanic plumes. Geochimica Et Cosmochimica Acta, 70, 2935-2941.
- Pandis, S.N., and Seinfeld, J.H., 1989. Sensitivity analysis of a chemical mechanism for aqueous-phase atmospheric chemistry. Journal of Geophysical Research, 94D, 1105-1126.
- Posch, M., and Reinds, G.J. 2009. A very simple dynamic soil acidification model for scenario analyses and target load calculations. Environmental Modelling & Software, 24, 329-340.

- Pyle, D.M., and Mather, T.A., 2009. Halogens in igneous processes and their fluxes to the atmosphere and oceans from volcanic activity: A review. Chemical Geology, 263, 110-121.
- Redington, A.L., Derwent, R.G., Ryall, D.B., Matthew, S., and Manning, A.J., 2001. Pollution of the Urban Midlands Atmosphere: Development of an 'urban Airshed' Model for the West Midlands. Hadley Centre Technical Note 31. Met Office, Bracknell. http://www.metoffice.gov.uk/research/hadleycentre/pubs/
- Redington A.L., Derwent R.G., Witham C.S. and Manning A.J., 2009, Sensitivity of modelled sulphate and nitrate aerosol to cloud, pH and ammonia emissions, Atmospheric Environment, 43, 3227-3234.
- Roberts, T.J. et al., 2009. Modelling reactive halogen formation and ozone depletion in volcanic plumes. Chemical Geology, 1-4, 151-163.
- Rose, W.I. et al., 2006. Atmospheric chemistry of a 33-34 hour old volcanic cloud from Hekla Volcano (Iceland): Insights from direct sampling and the application of chemical box modeling. Journal of Geophysical Research-Atmospheres, 111.
- RoTAP, 2012. Review of Transboundary Air Pollution: Acidifcation, Eutrophication, Ground Level Ozone and Heavy Metals in the UK. Contract Report to the Department for Environment, Food and Rural Affairs. Centre for Ecology and Hydrology. www.rotap.ceh.ac.uk
- Sawyer, G.M. et al., 2011. Gas and aerosol emissions from Villarrica volcano, Chile. Journal of Volcanology and Geothermal Research, 203, 62-75.
- Schmidt, A., Ostro, B., Carslaw, K.A., Wilson, M., Thordarson, T., Mann, G.W., and Simmons, A.J., 2011. Excess mortality in Europe following a future Laki-style Icelandic eruption. PNAS, 108 (38), 15710-15715.
- Schmidt, A., Thordarson, T., Oman, L.D., Robock, A., and Self, S., 2012. Climatic impact of the long-lasting 1783 Laki eruption: Inapplicability of massindependent sulfur isotopic composition measurements. Journal of Geophysical Research, 117, D23116, doi:10.1029/2012JD018414
- Schmidt, A., Witham, C.S., Richards, N.A.D., Thordarson, T., Theys, N., Feng, W.,
   Szpek, K., Johnson, B.T., Woolley, A.M., Jones, A.R., Redington, A.L., Hort,
   M.C., and Carslaw, K.S., 2014. Assessing hazards to aviation from sulfur
   dioxide emitted by explosive Icelandic eruptions, Journal of Geophysical
   Research Atmospheres, 119(24), doi:10.1002/2014JD022070.
- Shinohara, H., 2013. Volatile flux from subduction zone volcanoes: Insights from a detailed evaluation of the fluxes from volcanoes in Japan. Journal of Volcanology and Geothermal Research, 268, 46-63.
- Shinohara, H. and Witter, J.B., 2005. Volcanic gases emitted during mild Strombolian activity of Villarrica volcano, Chile. Geophysical Research Letters, 32.
- Simpson, D., Benedictow, A., Berge, H., Bergström, R., Emberson, L. D., Fagerli, H., Hayman, G. D., Gauss, M., Jonson, J. E., Jenkin, M. E., Nyíri, A., Richter, C., Semeena, V. S., Tsyro, S., Tuovinen, J. P., Valdebenito, Á., and Wind, P., 2012. The EMEP MSC-W chemical transport model; Part 1: Model description, Atmospheric Chemistry and Physics Discussions, 12, 3781-3874, 10.5194/acpd-12-3781-2012.
- Skeffington, R., Hall, J., Heywood, E., Wadsworth, R.A., Whitehead, P., Reynolds, B., Abbott, J., and Vincent, K., 2007. Uncertainty in critical load assessment models. Final report to the Environment Agency. R&D Project no. P4-120/4: Acidification and Annual Audits. March 2007. 152 pages. SC030172/SR.
- Stevenson, D.S., Johnson, C.E., Highwood, E.J., Gauci, V., Colins, W.J., and Derwent, R.G., 2003. Atmospheric impact of the 1783–1784 Laki eruption: Part I. Chemistry modelling. Atmospheric Chemistry and Physics, 3, 551–596.
- Symonds, R.B., Rose, W.I., Bluth, G.J.S., and Gerlach, T.M., 1994. Volcanic-gas studies: methods, results and applications. in Volatiles in Magmas, Reviews of Mineralogy, Vol. 30 (eds. Carroll, M.R. & Holloway, J.R.) 1–66.

- Tang, Y.S, Poskitt, J., Cape, J.N., Nemitz, E., Bealey, W.J., Leaver, D., Beith, S., Thacker, S., Simmons, I., Letho, K., Wood, C., Pereira, G., Sutton, M.A., Davies, M., Conolly, C., Donovan, B., and Braban, C.F., 2013. UK Eutrophying and Acidifying Atmospheric Pollutant project's Acid Gas and Aerosol Network (Data funded by Defra and the Devolved Administrations and published under the Open Government Licence v1.0, AGANet, http://ukair.defra.gov.uk/networks/network-info?view=ukeap), Date received: July 2013.
- Thordarson, T., and Self, S., 1993. The Laki (Skaftár Fires) and Grímsvötn eruptions in 1783–1785. Bulletin of Volcanology, 55, 233–263.
- Thordarson, T., and Self, S., 2003. Atmospheric and environmental effects of the 1783-1784 Laki eruption: A review and reassessment. Journal of Geophysical Research Atmospheres, 108 (1), pp. AAC 7-1 AAC 7-29.
- Vieno, M., Dore, A. J., Stevenson, D. S., Doherty, R., Heal, M. R., Reis, S., Hallsworth, S., Tarrason, L., Wind, P., Fowler, D., Simpson, D., and Sutton, M. A., 2010. Modelling surface ozone during the 2003 heat-wave in the UK, Atmospheric Chemistry and Physics, 10, 7963-7978, DOI 10.5194/acp-10-7963-2010.
- Vieno, M., Heal, M. R., Hallsworth, S., Famulari, D., Doherty, R. M., Dore, A. J., Tang, Y. S., Braban, C. F., Leaver, D., Sutton, M. A., and Reis, S., 2014. The role of long-range transport and domestic emissions in determining atmospheric secondary inorganic particle concentrations across the UK, Atmospheric Chemistry and Physics, 14, 8435-8447.
- von Glasow, R., Bobrowski, N., and Kern, C., 2009. The effects of volcanic eruptions on atmospheric chemistry. Chemical Geology, 263, 131-142.
- Werner, C., Kelly, P.J., Doukas, M., Lopez, T., Pfeffer, M., McGimsey, R., and Neal, C., 2013. Degassing of CO2, SO2, and H2S associated with the 2009 eruption of Redoubt Volcano, Alaska. Journal of Volcanology and Geothermal Research 259, 270-284.
- Witter, J.B., Kress, V.C., Delmelle, P. and Stix, J., 2004. Volatile degassing, petrology, and magma dynamics of the Villarrica Lava Lake, Southern Chile. Journal of Volcanology and Geothermal Research, 134, 303-337.

# Annex A: Communication Activities Related to this Project

The Project team conducted a number of activities to promote and discuss the work:

- The project was highlighted in a press release from BGS in August 2013 when the 'Source Characterisation' report was made available online (http://www.bgs.ac.uk/research/volcanoes/LakiEruptionScenarioPlanning.html). Subsequent press interviews were given by BGS.
- The project was presented (poster) at the second Volcano Observatories Best Practice (VOBP) workshop on 'Communication' held in Erice, Italy on 6-10 November 2013 and was discussed in the context of the Hyogo Framework for Action (HFA) by Sue Loughlin (workshop co-organiser). This workshop was attended by ~70 representatives of volcano observatories worldwide and academics from key institutions such as BGS, USGS, INGV.
- The project was mentioned in presentations at the WMO-IUGG "Ash dispersal forecast and civil aviation" workshop in Geneva on 18-20 Nov 2013, attended by project members Claire Witham (MO), Sue Loughlin (BGS and one of the meeting co-organisers), Anja Schmidt (Leeds). This workshop was attended by ~100 scientists and stakeholders.
- Press interviews on science efforts towards UK planning and preparedness were given in January 2014 during the 50<sup>th</sup> Anniversary 'Volcanic and Magmatic Studies Group meeting' in Edinburgh. The VOBP poster was on display at a BGS stand throughout the meeting.
- The project was mentioned during the VANAHEIM project stakeholders meeting on 18 March 2014, which was attended by CCS.
- A joint abstract was submitted to the EGU 2014 meeting by the project partners and CCS titled "UK Hazard Assessment for a Laki-type Volcanic Eruption". Claire Witham presented the work in a talk in the "Quantifying Volcanic Hazards" session on 28 April 2014. The abstract was picked up on by the press in the weekend preceding the meeting resulting in a mixture of largely misrepresentative articles. Claire gave an interview to the BBC whilst at EGU and the subsequent article was more balanced (http://www.bbc.co.uk/news/science-environment-27239321).
- In response to some of the interest and media associated with the EGU abstract, Anja Schmidt wrote a guest article for the Climate And Geohazards blog (http://climateandgeohazards.wordpress.com/2014/04/05/guest-blog-manmade-vs-volcanic-air-pollution/) which also appeared on the Natural History Museum's London Volcano Exhibit website (http://londonvolcano.com/2014/05/30/guest-blog-man-made-vs-volcanic-airpollution/).
- An abstract on the work was submitted to the Cities on Volcanoes 8 conference in Sep 2014.

# Annex B: Near surface model results

## Maps of probability of exceeding thresholds

Maps of probability of exceedance of AQI thresholds at the surface for daily and for hourly means. Maps are for the probability of being greater than the index concentration stated. Figures B-1 to B-4 give the probability that the specified threshold is exceeded at any point in a 6-week eruption period, based on the 80 eruption simulations. In all figures, areas coloured white do not experience concentrations above the given level at any time during any of the 80 simulation periods, hence the probability is zero.

Figure B-1: Percentage probability that at any time in a 6-week eruption the daily SO<sub>2</sub> concentrations will exceed the given AQI threshold concentrations for each grid square.



Figure B-2: Percentage probability that at any time in a 6-week eruption the hourly SO<sub>2</sub> concentrations will exceed the given AQI threshold concentrations for each grid square.

NAME

EMEP4UK



Figure B-3: Percentage probability that at any time in a 6-week eruption the daily SO<sub>4</sub> concentrations will exceed the given AQI threshold concentrations for each grid square.



Figure B-4: Percentage probability that at any time in a 6-week eruption the hourly SO<sub>4</sub> concentrations will exceed the given AQI threshold concentrations for each grid square.





## Maps of consecutive duration of exceedances

Maps of the maximum number of consecutive days/hours of exceedance of each threshold at the surface for daily and for hourly averages. This is the maximum consecutive duration modelled in any of the 80 simulated periods for each grid square.

Figure B-5: Maximum number of consecutive days that modelled SO<sub>2</sub> concentrations are above the given AQI threshold concentrations. There are no consecutive days above  $178 \ \mu g/m^3$ .



Figure B-6: Maximum number of consecutive hours that modelled  $SO_2$  concentrations are above the given AQI threshold concentrations. There are no consecutive hours above 1607  $\mu$ g/m<sup>3</sup>.



Figure B-7: Maximum number of consecutive days that modelled SO<sub>4</sub> concentrations are above the given AQI threshold concentrations.



Figure B-8: Maximum number of consecutive hours that modelled SO<sub>4</sub> concentrations are above the given AQI threshold concentrations.



## Maps of mean total time thresholds are exceeded

Maps of the mean number of hours and days (24-hour mean) a certain threshold could be exceeded at the surface per 6-week period. These are derived from the total number of exceeded hours and days in the whole 80 simulation periods divided by 80.

Figure B-9: Mean number of days in an eruption period that modelled SO<sub>2</sub> concentrations are above the given AQI threshold concentrations.



Figure B-10: Mean number of hours (>1 hour) in an eruption period that modelled  $SO_2$  concentrations are above the given AQI threshold concentrations. There are no values greater than 1 for the other thresholds.



Figure B-11: Mean number of days in an eruption period that modelled SO<sub>4</sub> concentrations are above the given AQI threshold concentrations. Note different plotting colour scales to SO<sub>2</sub>.





Figure B-12: Mean number of hours (>1 hour) in an eruption period that modelled SO<sub>4</sub> concentrations are above the given AQI threshold concentrations.



# Annex C: Flight level model results

## Maps of probability of exceeding thresholds

Maps of probability of exceedance of AQI thresholds at flight levels for hourly means. Maps are for the probability of being greater than the index concentration stated.

## $SO_2$



Figure C-1: Percentage probability that at any time in a 6-week eruption period hourly  $SO_2$  concentrations will exceed the given AQI threshold concentrations for each grid square at FL010.



Figure C-2: Percentage probability that at any time in a 6-week eruption period hourly SO<sub>2</sub> concentrations will exceed the given AQI threshold concentrations for each grid square at FL070.



Figure C-3: Percentage probability that at any time in a 6-week eruption period hourly SO<sub>2</sub> concentrations will exceed the given AQI threshold concentrations for each grid square at FL150.



Figure C-4: Percentage probability that at any time in a 6-week eruption period hourly SO<sub>2</sub> concentrations will exceed the given AQI threshold concentrations for each grid square at FL240.



Figure C-5: Percentage probability that at any time in a 6-week eruption period hourly SO<sub>2</sub> concentrations will exceed the given AQI threshold concentrations for each grid square at FL300.



Figure C-6: Percentage probability that at any time in a 6-week eruption period hourly  $SO_2$  concentrations will exceed the given AQI threshold concentrations for each grid square at FL370.

SO<sub>4</sub>



Figure C-7: Percentage probability that at any time in a 6-week eruption period hourly SO<sub>4</sub> concentrations will exceed the given AQI threshold concentrations for each grid square at FL010. White areas in the NAME outputs relate to FL010 not being a valid height over high topography, some areas are also zero probability above 151  $\mu$ g/m<sup>3</sup>.


Figure C-8: Percentage probability that at any time in a 6-week eruption period hourly SO<sub>4</sub> concentrations will exceed the given AQI threshold concentrations for each grid square at FL070.



Figure C-9: Percentage probability that at any time in a 6-week eruption period hourly  $SO_4$  concentrations will exceed the given AQI threshold concentrations for each grid square at FL150.



Figure C-10: Percentage probability that at any time in a 6-week eruption period hourly SO<sub>4</sub> concentrations will exceed the given AQI threshold concentrations for each grid square at FL240.



Figure C-11: Percentage probability that at any time in a 6-week eruption period hourly SO<sub>4</sub> concentrations will exceed the given AQI threshold concentrations for each grid square at FL300.



Figure C-12: Percentage probability that at any time in a 6-week eruption period hourly SO<sub>4</sub> concentrations will exceed the given AQI threshold concentrations for each grid square at FL370.

## Maps of duration of consecutive exceedances

Maps of the maximum number of consecutive hours number of hours each AQI threshold from Moderate4 could be exceeded at each flight level

## SO<sub>2</sub>



Figure C-13: Maximum number of consecutive hours that modelled  $SO_2$  concentrations are above the given AQI threshold concentrations at FL010.



Figure C-14: Maximum number of consecutive hours that modelled SO<sub>2</sub> concentrations are above the given AQI threshold concentrations at FL070.



Figure C-15: Maximum number of consecutive hours that modelled  $SO_2$  concentrations are above the given AQI threshold concentrations at FL150.



Figure C-16: Maximum number of consecutive hours that modelled  $SO_2$  concentrations are above the given AQI threshold concentrations at FL240.



Figure C-17: Maximum number of consecutive hours that modelled SO<sub>2</sub> concentrations are above the given AQI threshold concentrations at FL300.



Figure C-18: Maximum number of consecutive hours that modelled SO<sub>2</sub> concentrations are above the given AQI threshold concentrations at FL370.

 $SO_4$ 



Figure C-19: Maximum number of consecutive hours that modelled SO<sub>4</sub> concentrations are above the given AQI threshold concentrations at FL010.



Figure C-20: Maximum number of consecutive hours that modelled SO<sub>4</sub> concentrations are above the given AQI threshold concentrations at FL070.



Figure C-21: Maximum number of consecutive hours that modelled  $SO_4$  concentrations are above the given AQI threshold concentrations at FL150.



Figure C-22: Maximum number of consecutive hours that modelled  $SO_4$  concentrations are above the given AQI threshold concentrations at FL240.



Figure C-23: Maximum number of consecutive hours that modelled SO<sub>4</sub> concentrations are above the given AQI threshold concentrations at FL300.



Figure C-24: Maximum number of consecutive hours that modelled SO<sub>4</sub> concentrations are above the given AQI threshold concentrations at FL370.

### Maps of total time thresholds are exceeded

Maps of the mean number of hours each AQI threshold from Moderate4 could be exceeded at each flight level in a 6-week eruption period. These are derived from the total number of exceeded hours and days in the whole 80 simulation periods divided by 80.

 $SO_2$ 

## FL010

NAME No exceedances greater than an hour above the Moderate4 level.

**EMEP4UK** No exceedances greater than an hour above the Moderate4 level.

Figure C-25: Mean number of hours in an eruption period that modelled SO<sub>2</sub> concentrations are above the given AQI threshold concentrations at FL010.

## FL070

#### NAME





Figure C-26: Mean number of hours in an eruption period that modelled  $SO_2$  concentrations are above the given AQI threshold concentrations at FL070.



Figure C-27: Mean number of hours in an eruption period that modelled  $SO_2$  concentrations are above the given AQI threshold concentrations at FL150. (Note: white values over Iceland are >54 hours).



Figure C-28: Mean number of hours in an eruption period that modelled  $SO_2$  concentrations are above the given AQI threshold concentrations at FL240. (Note: white values over Iceland are >54 hours).



Figure C-29: Mean number of hours in an eruption period that modelled  $SO_2$  concentrations are above the given AQI threshold concentrations at FL300. (Note: white values over Iceland are >54 hours).



Figure C-30: Mean number of hours in an eruption period that modelled SO<sub>2</sub> concentrations are above the given AQI threshold concentrations at FL370. Note: different contour scale has been used to other flight levels to account for larger total times at FL370.

 $SO_4$ 

NAME

FL010

EMEP4UK



Figure C-31: Mean number of hours in an eruption period that modelled  $SO_4$  concentrations are above the given AQI threshold concentrations at FL010.



Figure C32: Mean number of hours in an eruption period that modelled  $SO_4$  concentrations are above the given AQI threshold concentrations at FL070.



Figure C-33: Mean number of hours in an eruption period that modelled  $SO_4$  concentrations are above the given AQI threshold concentrations at FL1500.



Figure C-34: Mean number of hours in an eruption period that modelled SO<sub>4</sub> concentrations are above the given AQI threshold concentrations at FL240.



Figure C-35: Mean number of hours in an eruption period that modelled  $SO_4$  concentrations are above the given AQI threshold concentrations at FL300.

FL370

#### NAME

No exceedances above Moderate4

No exceedances above Moderate4

EMEP4UK

Figure C-36: Mean number of hours in an eruption period that modelled  $SO_4$  concentrations are above the given AQI threshold concentrations at FL370.

# Annex D: Sensitivity tests

## Near-surface

For each simulation maps are presented for:

- The maximum hourly value simulated at each grid square at any time during the simulation (these maxima do not necessarily occur at the same time in each grid square)
- The hourly snap-shot values equivalent to those depicted in Figure 1. For SO<sub>2</sub> this is the output at 18:00UTC 27/06/2010 and for SO<sub>4</sub> at 03:00 01/07/2010.



Figure D-1: Maximum values for sensitivity test M1.



Figure D-2: Maximum values for sensitivity test M2.



Figure D-3: Maximum values for sensitivity test H1.



Figure D-4: Maximum values for sensitivity test H2.



Figure D-5: Maximum values for sensitivity test T1.



Figure D-6: Maximum values for sensitivity test T2.

## Flight level (FL300-FL325)

Maps are not given below for M1 and M2 as the evolution of the plume is the same as the initial simulation in terms of its shape and dispersion. In these scenarios, only the magnitude of the concentration varies and this is approximately linear with the amount of mass initially released.



Figure D-7: Time evolution of the plume in the initial and H1 sensitivity tests.



Figure D-8: Time evolution of the plume in the T1 and T2 sensitivity tests.
# Annex E: Acidity critical load exceedance statistics by habitat and country

# (i) Baseline scenario

# Results for England:

						Average
			Exceeded	Percentage	Accumulated	Accumulated
	EUNIS	Habitat	Area	Area	Exceedance	Exceedance
Broad Habitat	class(es)	Area (km <sup>2</sup> )	(km²)	Exceeded	(keq/year)	(keq/ha/year)
Acid grassland	E1.7 & E3.52	2669	1506	56.4	45925	0.17
Calcareous grassland	E1.26	1714	0	0.0	0	0
Dwarf shrub heath	F4.11 & F4.2	2462	999	40.6	19555	0.08
Bog	D1	1006	776	77.2	26609	0.26
Montane	E4.2	2	1	75.0	36	0.19
Coniferous woodland (managed)	G3	1716	732	42.7	43620	0.25
Broadleaved woodland (managed)	G1	5565	748	13.4	36294	0.07
Unmanaged woods	G4	2392	202	8.4	7513	0.03
Freshwaters	C1 & C2	1109	291	26.3	12849	0.12
All habitats		18635	5255	28.2	192401	0.1

## Results for Wales:

						Average
			Exceeded	Percentage	Accumulated	Accumulated
	EUNIS	Habitat	Area	Area	Exceedance	Exceedance
Broad Habitat	class(es)	Area (km <sup>2</sup> )	(km²)	Exceeded	(keq/year)	(keq/ha/year)
Acid grassland	E1.7 & E3.52	3143	1832	58.3	61368	0.2
Calcareous grassland	E1.26	45	0	0.0	0	0
Dwarf shrub heath	F4.11 & F4.2	1078	316	29.3	6642	0.06
Bog	D1	56	26	46.9	695	0.12
Montane	E4.2	18	17	97.2	917	0.51
Coniferous woodland (managed)	G3	1048	378	36.0	13400	0.13
Broadleaved woodland (managed)	G1	790	168	21.2	6516	0.08
Unmanaged woods	G4	395	66	16.7	2400	0.06
Freshwaters	C1 & C2	1225	147	12.0	5926	0.05
All habitats		7798	2950	37.8	97864	0.13

#### Results for Scotland:

						Average
			Exceeded	Percentage	Accumulated	Accumulated
	EUNIS	Habitat	Area	Area	Exceedance	Exceedance
Broad Habitat	class(es)	Area (km <sup>2</sup> )	(km <sup>2</sup> )	Exceeded	(keq/year)	(keq/ha/year)
Acid grassland	E1.7 & E3.52	8336	2730	32.8	79349	0.1
Calcareous grassland	E1.26	7	0	0.0	0	0
Dwarf shrub heath	F4.11 & F4.2	20190	2439	12.1	52516	0.03
Bog	D1	3955	558	14.1	10892	0.03
Montane	E4.2	3034	1440	47.5	49481	0.16
Coniferous woodland (managed)	G3	5111	821	16.1	23066	0.05
Broadleaved woodland (managed)	G1	1096	164	15.0	5465	0.05
Unmanaged woods	G4	1016	95	9.3	2773	0.03
Freshwaters	C1 & C2	5338	228	4.3	5155	0.01
All habitats		48083	8475	17.6	228697	0.05

# Results for Northern Ireland:

						Average
			Exceeded	Percentage	Accumulated	Accumulated
	EUNIS	Habitat	Area	Area	Exceedance	Exceedance
Broad Habitat	class(es)	Area (km <sup>2</sup> )	(km²)	Exceeded	(keq/year)	(keq/ha/year)
Acid grassland	E1.7 & E3.52	1189	359	30.2	10184	0.09
Calcareous grassland	E1.26	43	0	0.0	0	0
Dwarf shrub heath	F4.11 & F4.2	974	92	9.4	2444	0.03
Bog	D1	437	163	37.4	2969	0.07
Montane	E4.2	0	0	0.0	0	0
Coniferous woodland (managed)	G3	500	60	12.0	1251	0.03
Broadleaved woodland (managed)	G1	0	0	0.0	0	0
Unmanaged woods	G4	208	16	7.9	623	0.03
Freshwaters	C1 & C2	186	12	6.6	496	0.03
All habitats		3537	702	19.8	17967	0.05

## Results for the UK:

						Average
			Exceeded	Percentage	Accumulated	Accumulated
	EUNIS	Habitat	Area	Area	Exceedance	Exceedance
Broad Habitat	class(es)	Area (km <sup>2</sup> )	(km²)	Exceeded	(keq/year)	(keq/ha/year)
Acid grassland	E1.7 & E3.52	15336	6428	41.9	196826	0.13
Calcareous grassland	E1.26	1808	0	0.0	0	0
Dwarf shrub heath	F4.11 & F4.2	24705	3847	15.6	81157	0.03
Bog	D1	5454	1524	27.9	41165	0.08
Montane	E4.2	3054	1459	47.8	50434	0.17
Coniferous woodland (managed)	G3	8374	1991	23.8	81336	0.1
Broadleaved woodland (managed)	G1	7452	1080	14.5	48275	0.06
Unmanaged woods	G4	4011	379	9.4	13310	0.03
Freshwaters	C1 & C2	7857	678	8.6	24425	0.03
All habitats		78051	17386	22.3	536928	0.07

## Notes:

- a) EUNIS classes are the classes of the European Nature Information System; a hierarchical habitat classification system (ref) used when submitting UK critical loads data to Europe for work under the UNECE Convention on Long-Range Transboundary Air Pollution. The EUNIS classes shown are those that most closely correspond to the UK broad habitat types listed and mapped for critical loads research in the UK.
- b) Managed woodland is assumed to be primarily productive forest where harvesting and removal of trees takes place, whereas "unmanaged woodland" is assumed to be "managed" only for biodiversity or amenity rather than timber production.
- c) The habitat area for freshwaters is based on the catchment areas of 1752 selected lake or stream catchments, mainly in upland, acid-sensitive regions of the UK.
- d) The habitat areas used for UK critical loads research (a) only include areas where data exist for the calculation or derivation of critical loads; (b) may differ from other national habitat distribution maps or estimates of habitat areas.

# (ii) Average Laki-type scenario

# Results for England:

						Average
				Percentage	Accumulated	Accumulated
	EUNIS	Habitat Area	Exceeded	Area	Exceedance	Exceedance
Broad Habitat	class(es)	(km²)	Area (km <sup>2</sup> )	Exceeded	(keq/year)	(keq/ha/year)
Acid grassland	E1.7 & E3.52	2669	1890	70.8	70771	0.27
Calcareous grassland	E1.26	1714	0	0.0	0	0
Dwarf shrub heath	F4.11 & F4.2	2462	1446	58.7	32657	0.13
Bog	D1	1006	888	88.3	37587	0.37
Montane	E4.2	2	2	95.8	73	0.38
Coniferous woodland (managed)	G3	1716	813	47.4	51056	0.3
Broadleaved woodland (managed)	G1	5565	830	14.9	43149	0.08
Unmanaged woods	G4	2392	238	9.9	9554	0.04
Freshwaters	C1 & C2	1109	332	30.0	16023	0.14
All habitats		18635	6439	34.6	260870	0.14

# Results for Wales:

						Average
				Percentage	Accumulated	Accumulated
	EUNIS	Habitat Area	Exceeded	Area	Exceedance	Exceedance
Broad Habitat	class(es)	(km²)	Area (km <sup>2</sup> )	Exceeded	(keq/year)	(keq/ha/year)
Acid grassland	E1.7 & E3.52	3143	2509	79.8	105419	0.34
Calcareous grassland	E1.26	45	0	0.0	0	0
Dwarf shrub heath	F4.11 & F4.2	1078	524	48.6	14235	0.13
Bog	D1	56	40	72.1	1416	0.25
Montane	E4.2	18	18	97.8	1400	0.78
Coniferous woodland (managed)	G3	1048	480	45.8	21326	0.2
Broadleaved woodland (managed)	G1	790	237	30.0	9909	0.13
Unmanaged woods	G4	395	99	25.0	3856	0.1
Freshwaters	C1 & C2	1225	192	15.7	10267	0.08
All habitats		7798	4099	52.6	167828	0.22

# Results for Scotland:

						Average
				Percentage	Accumulated	Accumulated
	EUNIS	Habitat Area	Exceeded	Area	Exceedance	Exceedance
Broad Habitat	class(es)	(km²)	Area (km <sup>2</sup> )	Exceeded	(keq/year)	(keq/ha/year)
Acid grassland	E1.7 & E3.52	8336	5832	70.0	237158	0.28
Calcareous grassland	E1.26	7	0	0.0	0	0
Dwarf shrub heath	F4.11 & F4.2	20190	14034	69.5	466523	0.23
Bog	D1	3955	2823	71.4	83489	0.21
Montane	E4.2	3034	2779	91.6	147993	0.49
Coniferous woodland (managed)	G3	5111	1455	28.5	45086	0.09
Broadleaved woodland (managed)	G1	1096	302	27.5	9907	0.09
Unmanaged woods	G4	1016	176	17.4	6210	0.06
Freshwaters	C1 & C2	5338	437	8.2	16162	0.03
All habitats		48083	27838	57.9	1012528	0.21

# Results for Northern Ireland:

						Average
				Percentage	Accumulated	Accumulated
	EUNIS	Habitat Area	Exceeded	Area	Exceedance	Exceedance
Broad Habitat	class(es)	(km²)	Area (km <sup>2</sup> )	Exceeded	(keq/year)	(keq/ha/year)
Acid grassland	E1.7 & E3.52	1189	612	51.5	19935	0.17
Calcareous grassland	E1.26	43	0	0.0	0	0
Dwarf shrub heath	F4.11 & F4.2	974	308	31.6	6779	0.07
Bog	D1	437	313	71.7	8561	0.2
Montane	E4.2	0	0	0.0	0	0
Coniferous woodland (managed)	G3	500	131	26.2	3764	0.08
Broadleaved woodland (managed)	G1	0	0	0.0	0	0
Unmanaged woods	G4	208	20	9.5	905	0.04
Freshwaters	C1 & C2	186	24	12.9	706	0.04
All habitats		3537	1408	39.8	40650	0.11

#### Results for UK:

						Average
				Percentage	Accumulated	Accumulated
	EUNIS	Habitat Area	Exceeded	Area	Exceedance	Exceedance
Broad Habitat	class(es)	(km²)	Area (km <sup>2</sup> )	Exceeded	(keq/year)	(keq/ha/year)
Acid grassland	E1.7 & E3.52	15336	10843	70.7	433282	0.28
Calcareous grassland	E1.26	1808	0	0.0	0	0
Dwarf shrub heath	F4.11 & F4.2	24705	16312	66.0	520193	0.21
Bog	D1	5454	4065	74.5	131053	0.24
Montane	E4.2	3054	2799	91.6	149466	0.49
Coniferous woodland (managed)	G3	8374	2880	34.4	121231	0.14
Broadleaved woodland (managed)	G1	7452	1369	18.4	62965	0.08
Unmanaged woods	G4	4011	533	13.3	20525	0.05
Freshwaters	C1 & C2	7857	985	12.5	43158	0.05
All habitats		78051	39786	51.0	1481873	0.19

#### Notes:

- e) EUNIS classes are the classes of the European Nature Information System; a hierarchical habitat classification system (ref) used when submitting UK critical loads data to Europe for work under the UNECE Convention on Long-Range Transboundary Air Pollution. The EUNIS classes shown are those that most closely correspond to the UK broad habitat types listed and mapped for critical loads research in the UK.
- f) Managed woodland is assumed to be primarily productive forest where harvesting and removal of trees takes place, whereas "unmanaged woodland" is assumed to be "managed" only for biodiversity or amenity rather than timber production.
- g) The habitat area for freshwaters is based on the catchment areas of 1752 selected lake or stream catchments, mainly in upland, acid-sensitive regions of the UK.
- h) The habitat areas used for UK critical loads research (a) only include areas where data exist for the calculation or derivation of critical loads; (b) may differ from other national habitat distribution maps or estimates of habitat areas.

# Annex F: Literature on HCI, HF and H<sub>2</sub>S observations

Volcano	Country	Magma type	Method	SO2/HCI	HCI/SO2	SO2/HF	HF/SO2	Reference
Ambrym	Vanuatu	Basalt	FP	4.3	0.23	7.9	0.13	Aiuppa (2009)
(Bembrow)								
Ambrym (Mbelesu)	Vanuatu	Basalt	FP	4.7	0.21	10.7	0.09	Aiuppa (2009)
Asama	Japan		GS	5	0.20			Shinohara (2013)
Aso	Japan		GS	14.29	0.07			Shinohara (2013)
Aso	Japan		FP	100	0.01	33.3	0.03	Roberts (2009)
Aso	Japan		FP	10	0.10			Roberts (2009)
Augustine	USA	Andesite	DS	0.7	1.43	26	0.04	Aiuppa (2009)
Cerro Negro	Mexico	Basalt	DS	2.1	0.48	15.2	0.07	Aiuppa (2009)
Colima	Mexico	Andesite	DS	3.9	0.26	59	0.02	Aiuppa (2009)
Erebus	Antarctica	Anorthoclase phonolite	FP	0.6	1.67	0.7	1.43	Aiuppa (2009)
Erta Ale	Ethiopia	Basalt	FP	2.6	0.38	2.9	0.34	Aiuppa (2009)
Etna	Italy	Hawaiite	FP/FTIR	2.4	0.42	6.9	0.14	Aiuppa (2009)
Eyjafjallajokull	Iceland	Basaltic/basaltic andesitic	L	0.94	1.06	34	0.03	Bagnato (2013)
Fuego	Guatemala	Basalt	FP	5.2	0.19	72	0.01	Aiuppa (2009)
Galunggung	Iceland	Basalt/basaltic andesite	Ash Leachate	4.4	0.23	29	0.03	Aiuppa (2009)
Hekla	lceland	Basaltic Icelandite	SS	18	0.06	10.7	0.09	Aiuppa (2009)
Helka	lceland	Basaltic Icelandite	CIMS	14.7	0.07			Rose (2006)
Kilauea	Hawaii, USA	Basalt	FP	26.3	0.04	28.6	0.03	Mather (2012)
Kilauea	Hawaii, USA	Basalt	FP	18.52	0.05	24.4	0.04	Mather (2012)
Kilauea	Hawaii, USA	Basalt	FP / DS	34	0.03	30	0.03	Aiuppa (2009)
Klyuchevshoi	Russia	Basalt	DS	0.08	12.50	0.9	1.11	Aiuppa (2009)
Kudryavy	Russia	Basalt Andesite	DS	3.6	0.28	46	0.02	Aiuppa (2009)
Lascar	Chile	Andesite	FP	1.63	0.61	1.92	0.52	Mather (2004)
Lascar	Chile	Andesite	FP	1.7	0.59	4.3	0.23	Aiuppa (2009)
Masaya	Nicaragua	Basalt	FTIR / FP	2.2	0.45	12.1	0.08	Aiuppa (2009)
Merapi	Indonesia	Andesite	DS	0.7	1.43	10	0.10	Aiuppa (2009)
Miyakejima	Japan	Basaltic	GS	14.29	0.07			Shinohara (2013)
Miyakejima	Japan	Basaltic	FP	6.6	0.15	85	0.01	Aiuppa (2009)
Momotombo	Nicaragua	Andesite	DS	1.4	0.71	69	0.01	Aiuppa (2009)
Nyiragongo	DR Congo	Basanite	FP/FTIR	13.1	0.08	11.3	0.09	Aiuppa (2009)
Pacaya	Guatemala	basalt	FP	1.7	0.59	7.7	0.13	Aiuppa (2009)
Poas	Costa Rica	Basalt to Dacite	DS	1.9	0.53	16.1	0.06	Aiuppa (2009)
Popocateptl	Mexico	Andesite	FTIR	3.7	0.27	12.3	0.08	Aiuppa (2009)
Satsuma- Iwojima	Japan	Basaltic	GS	1.67	0.60			Shinohara (2013)
Stromboli	Italy	Trachybasalt	DP/DT	2	0.50	10	0.10	Aiuppa (2009)
Suwanosejima	Japan		GS	10	0.10			Shinohara (2013)
Telica	Nicaragua	Basalt	FP	2.2	0.45	12.6	0.08	Aiuppa (2009)
Tolbachik	Kamchatka	Basalt	DS	0.13	7.69	4.29	0.23	Aiuppa (2009)
Villarica	Chile	Basaltic Andesite	FP	1.42	0.70			Sawyer (2011)
Villarica	Chile	Basaltic Andesite	FTIR	3	0.33			Sawyer (2011)
Villarica	Chile	Basaltic Andesite	FP	1.69	0.59	4.35	0.23	Mather (2004)
Villarica	Chile	Basaltic Andesite	<u>FP</u>	2.6	0.38	10	0.10	Aiuppa (2009)
Yasur	Vanuatu	Basalt Andesite	F۲	3./	0.27	10	0.10	Aluppa (2009)

Table F-1: HCl and HF ratios from the volcanological literature.

Volcano	Country	Magma	Method	SO2/H2S	6 H2S/SO2	Ref
La Fossa-Vulcano	Italy		FP	0.35	2.86	Aiuppa (2009)
Showashinzan	Japan	Andesite		0.39	2.56	Halmer (2002)
La Fossa-Vulcano	Italy		UVS	0.45	2.22	Aiuppa (2009)
Mt St Helens	USA	Dacite-andesite	·	0.53	1.89	Halmer (2002)
Shinmoedake, Kirishima	Japan		Multi- GAS	0.8	1.25	Shinohara (2013)
Shinmoedake, Kirishima	Japan		Multi- GAS	0.8	1.25	Shinohara (2013)
Usu	Japan	Dacite-andesite		0.89	1.12	Halmer (2002)
La Fossa-Vulcano	Italy		FP	0.97	1.03	Aiuppa (2009)
Gunung, Merapi	Indonesia	Andesite		1.08	0.93	Halmer (2002)
Tolbachik, Kamchatka	Russia	Basalt		1.12	0.89	Halmer (2002)
Nyiragongo	DR Congo	Melilite-nephelinite		1.38	0.72	Halmer (2002)
Ngauruhoe	New Zealand	Andesite		1.5	0.67	Halmer (2002)
Momotombo	Nicaragua	Tholeitic-basalt		1.78	0.56	Halmer (2002)
La Fossa-Vulcano	Italy		UVS	2.1	0.48	Aiuppa (2009)
White Island	New Zealand	Andesite		2.2	0.45	Halmer (2002)
Shinmoedake, Kirishima	Japan		Multi- GAS	3.3	0.30	Shinohara (2013)
Satsuma-Iwojima	Japan	Basaltic	GS	5	0.20	Shinohara (2013)
Asama	Japan		GS	6.67	0.15	Shinohara (2013)
Etna	Italy	Hawaiite		7.31	0.14	Halmer (2002)
Ardoukoba	Djibouti	Tholeitic-basalt		8.38	0.12	Halmer (2002)
Erta'Ale	Ethiopia	Tholeitic-basalt		10.2	0.10	Halmer (2002)
Redoubt	USA	Andesite	IR	13.5	0.07	Werner (2013)
Miyakejima	Japan	Basaltic	GS	14.29	0.07	Shinohara (2013)
Aso	Japan		GS	20	0.05	Shinohara (2013)
Suwanosejima	Japan		GS	25	0.04	Shinohara (2013)
Poas	Costa Rica	Tholeitic-basalt		27.2	0.04	Halmer (2002)
Surtsey	Iceland	Alkali-basalt		27.3	0.04	Halmer (2002)
Kilauea	Hawaii, USA	Tholeitic-basalt		132.8	0.01	Halmer (2002)

Table F-2:  $H_2S$  ratios from the volcanological literature.

	Mean	St Dev	Median	Maximum	P90	P95	P99
Rosemaund	0.25	0.43	0.11	6.16	0.63	0.94	1.98
Narberth	0.23	0.40	0.10	5.48	0.58	0.89	2.06
Halladale	0.20	0.32	0.10	5.55	0.50	0.74	1.48
Auchencorth	0.22	0.35	0.10	5.02	0.53	0.81	1.63
Shetland	0.21	0.36	0.10	5.86	0.50	0.76	1.73
Glensaugh	0.22	0.36	0.10	5.01	0.52	0.81	1.72
Moor House	0.22	0.36	0.10	5.73	0.54	0.81	1.69
Rothamstead	0.26	0.43	0.12	7.05	0.64	0.96	2.19
Strathvaich Dam	0.20	0.33	0.10	4.20	0.51	0.75	1.60
Eskdalemuir	0.22	0.35	0.10	5.40	0.54	0.80	1.75
High Muffles	0.23	0.37	0.11	4.89	0.56	0.86	1.86
Stoke Ferry	0.26	0.43	0.11	6.19	0.61	0.96	2.28
Yarner Wood	0.23	0.40	0.10	5.76	0.57	0.86	1.97
Cromwell Road	0.25	0.41	0.12	4.62	0.63	0.95	2.20
Sutton Bonington	0.25	0.42	0.11	6.03	0.62	0.97	2.13
Lagganlia	0.21	0.34	0.10	6.96	0.51	0.78	1.68
Hillsborough	0.21	0.36	0.09	6.63	0.52	0.81	1.79
Lough Navar	0.20	0.35	0.09	6.03	0.52	0.76	1.57
Rum	0.21	0.35	0.10	7.15	0.52	0.76	1.67
Edinburgh	0.22	0.35	0.10	5.02	0.53	0.81	1.63
Cwmystwyth	0.23	0.40	0.10	5.88	0.58	0.86	1.92
Carradale	0.21	0.34	0.10	5.15	0.51	0.77	1.73
Barcombe Mills	0.25	0.39	0.12	4.52	0.60	0.91	1.92
Detling	0.26	0.42	0.12	4.78	0.61	0.93	2.37
Harwell	0.25	0.43	0.11	6.73	0.63	0.95	2.11
Ladybower	0.24	0.40	0.11	5.06	0.61	0.94	2.10
Plas_Y_Brenin	0.22	0.39	0.10	6.36	0.55	0.83	1.87
Caenby	0.24	0.40	0.11	5.68	0.58	0.94	2.06
Goonhilly	0.23	0.39	0.10	5.64	0.56	0.85	2.01

# Annex G: Summary statistics for H<sub>2</sub>S

Table G-1: Summary statistics for the daily average H<sub>2</sub>S modelled as Xinsol ( $X_{insol}$ :SO<sub>2</sub> = 0.04) concentrations over the 10 year dataset for thirty sites across the UK ( $\mu$ g/m<sup>3</sup>).

	Mean	St Dev	Median	Maximum	P90	P95	P99
Rosemaund	1.08	1.88	0.47	27.21	2.78	4.14	8.73
Narberth	1.02	1.79	0.43	24.19	2.56	3.95	9.11
Halladale	0.90	1.42	0.44	24.49	2.21	3.27	6.53
Auchencorth	0.95	1.55	0.44	22.18	2.32	3.59	7.21
Shetland	0.91	1.57	0.42	25.88	2.19	3.35	7.65
Glensaugh	0.97	1.58	0.46	22.12	2.30	3.57	7.57
Moor House	0.97	1.59	0.45	25.27	2.39	3.57	7.45
Rothamstead	1.13	1.91	0.51	31.12	2.82	4.25	9.66
Strathvaich Dam	0.90	1.45	0.43	18.56	2.24	3.32	7.08
Eskdalemuir	0.95	1.55	0.45	23.85	2.36	3.52	7.74
High_Muffles	1.01	1.63	0.47	21.58	2.47	3.82	8.20
Stoke Ferry	1.13	1.92	0.50	27.34	2.70	4.22	10.07
Yarner Wood	1.03	1.76	0.45	25.44	2.53	3.78	8.71
Cromwell Road	1.13	1.82	0.52	20.37	2.78	4.18	9.72
Sutton Bonington	1.11	1.87	0.48	26.62	2.74	4.28	9.42
Lagganlia	0.92	1.52	0.43	30.71	2.25	3.44	7.40
Hillsborough	0.93	1.60	0.41	29.25	2.28	3.57	7.89
Lough Navar	0.90	1.56	0.40	26.60	2.28	3.37	6.92
Rum	0.91	1.52	0.42	31.56	2.28	3.34	7.39
Edinburgh	0.95	1.55	0.44	22.18	2.32	3.59	7.21
Cwmystwyth	1.01	1.76	0.44	25.94	2.57	3.79	8.47
Carradale	0.91	1.49	0.42	22.71	2.27	3.38	7.63
Barcombe Mills	1.08	1.72	0.52	19.96	2.64	4.03	8.48
Detling	1.13	1.85	0.53	21.09	2.71	4.09	10.44
Harwell	1.12	1.91	0.50	29.72	2.78	4.21	9.29
Ladybower	1.07	1.77	0.46	22.32	2.68	4.15	9.29
Plas_Y_Brenin	0.98	1.71	0.43	28.07	2.43	3.66	8.26
Caenby	1.08	1.77	0.48	25.08	2.56	4.13	9.09
Goonhilly	1.01	1.71	0.43	24.90	2.45	3.75	8.86

Table G-2: Summary statistics for the daily average H2S modelled as Xinsol (Xinsol:SO2 = 0.3) concentrations over the 10 year dataset for thirty sites across the UK ( $\mu$ g/m<sup>3</sup>)

# Annex H: Site specific halogen results

Each of the figures in this section contains five time series graphs labelled A-E, which are as follows:

A: Volcano emissions (Tg);

B: Modelled (purple squares) and measured average ambient (orange line) HCI

C: Modelled SO<sub>2</sub>: Baseline (continuous), eruption scenario (crosses)

D: Modelled SO<sub>4</sub>: Baseline (continuous), eruption scenario (crosses)

E: Ratio of HCl to SO<sub>2</sub>: eruption scenario emission ratio (purple line); eruption scenariobaseline (green crosses).



Figure H-1: EMEP4UK 24 hour average results summary for London 2004.



Figure H-2: EMEP4UK 24 hour average results summary for Cromwell Road, London 2005.



Figure H-3: EMEP4UK 24 hour average results summary for Cromwell Road, London 2006.



Figure H-4:1 EMEP4UK 24 hour average results summary for Stoke Ferry 2003.



Figure H-5: EMEP4UK 24 hour average results summary for Stoke Ferry 2004.



Figure H-6: EMEP4UK 24 hour average results summary for Stoke Ferry 2005.



Figure H-7: EMEP4UK 24 hour average results summary for Stoke Ferry 2005.



Figure H-8: EMEP4UK 24 hour average results summary for Harwell 2003.



Figure H-9: EMEP4UK 24 hour average results summary for Harwell 2004.



Figure H-10: EMEP4UK 24 hour average results summary for Harwell 2005.



Figure H-11: EMEP4UK 24 hour average results summary for Harwell 2006.



Figure H-12: MEP4UK 24 hour average results summary for Goonhilly, Cornwall, 2003.



Figure H-13: EMEP4UK 24 hour average results summary for Goonhilly, Cornwall, 2004.



Figure H-14: EMEP4UK 24 hour average results summary for Goonhilly, Cornwall, 2005.



Figure H-15: EMEP4UK 24 hour average results summary for Goonhilly, Cornwall, 2006.



Figure H-16: EMEP4UK 24 hour average results summary for Auchencorth Moss, 2003.



Figure H-17: EMEP4UK 24 hour average results summary for Auchencorth Moss, Midlothian, 2004.



Figure H-18: EMEP4UK 24 hour average results summary for Auchencorth Moss, Midlothian, 2005.



Figure H-19: EMEP4UK 24 hour average results summary for Auchencorth Moss, Midlothian, 2005.



Figure H-20: EMEP4UK 24 hour average results summary for Shetland, 2003.



Figure H-21: EMEP4UK 24 hour average results summary for Shetland, Scotland, 2004.



Figure H-22: EMEP4UK 24 hour average results summary for Shetland, Scotland, 2005.



Figure H-23: EMEP4UK 24 hour average results summary for Shetland, Scotland, 2006.

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