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Understanding the environmental impacts of large fissure eruptions: Aerosol and gas emissions from the 2014–2015 Holuhraun eruption (Iceland)

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ABSTRACT

The 2014–2015 Holuhraun eruption in Iceland, emitted ~11 Tg of SO_2 into the troposphere over 6 months, and caused one of the most intense and widespread volcanogenic air pollution events in centuries. This study provides a number of source terms for characterisation of plumes in large fissure eruptions, in Iceland and elsewhere. We characterised the chemistry of aerosol particle matter (PM) and gas in the Holuhraun plume, and its evolution as the plume dispersed, both via measurements and modelling. The plume was sampled at the eruptive vent, and in two populated areas in Iceland. The plume caused repeated air pollution events, exceeding hourly air quality standards (350 μg/m^3) for SO_2 on 88 occasions in Reykjahlíð town (100 km distance), and 34 occasions in Reykjavík capital area (250 km distance). Average daily concentration of volcanogenic PM sulphate exceeded 5 μg/m^3 on 30 days in Reykjavík capital area, which is the maximum concentration measured during non-eruptive background interval. There are currently no established air quality standards for sulphate. Combining the results from direct sampling and dispersion modelling, we identified two types of plume impacting the downwind populated areas. The first type was characterised by high concentrations of both SO_2 and S-bearing PM, with a high S_{gas}/S_{PM} mass ratio ($SO_{2(g)}/SO_{4\ (PM)}^{2-} > 10$). The second type had a low S_{gas}/S_{PM} ratio (<10). We suggest that this second type was a mature plume where sulphur had undergone significant gas-to-aerosol conversion in the atmosphere. Both types of plume were rich in fine aerosol (predominantly PM_1 and $PM_{2.5}$), sulphate (on average ~90% of the PM mass) and various trace species, including heavy metals. The fine size of the volcanic PM mass (75–80% in $PM_{2.5}$), and the high environmental lability of its chemical components have potential adverse implications for environmental and health impacts. However, only the dispersion of volcanic SO_2 was forecast in public warnings and operationally monitored during the eruption. We make a recommendation that sulphur gas-to-aerosol conversion processes, and a sufficiently large model domain to contain the transport of a tropospheric plume on the timescale of days be utilized for public health and environmental impact forecasting in future eruptions in Iceland and elsewhere in the world.

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1. Introduction

Large fissure eruptions (>1 km^3) are one of the most environmentally hazardous volcanic scenarios due to their prodigious

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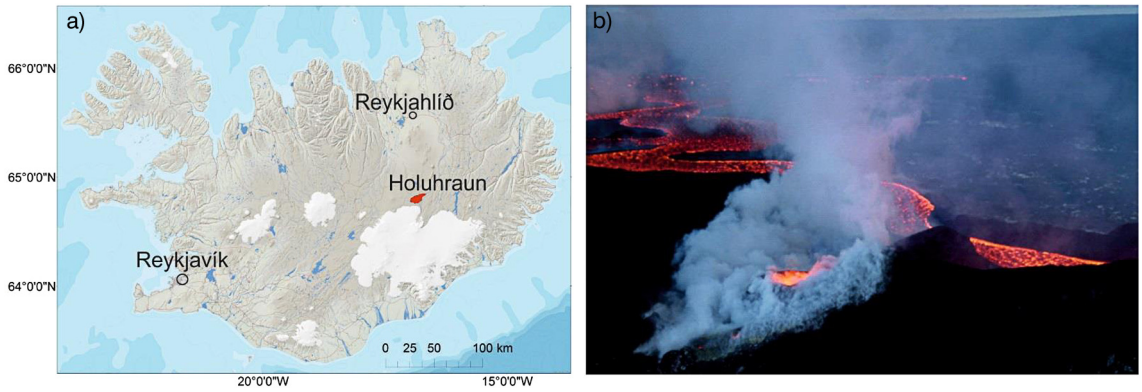

Fig. 1. a) Location map of the Holuhraun eruption site (lava flow extent shown by red colour), and the two populated areas in this study: Reykjavík city capital area, population 120,000; and Reykjahlíð town, population 300. **b**) Holuhraun eruption during the January 2015 field campaign. During this period the activity was exclusively effusive. The width of the lava flow is ∼50 m. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

release of gases and aerosol into the troposphere and lower stratosphere. This can impact air quality, terrestrial and aquatic environments, climate and health (Robock, 2000; Schmidt et al., 2011; Thordarson and Self, 2003; Witham et al., 2015). Icelandic large fissure eruptions were included in the UK National Risk Register in 2012 as one of the highest priority risks, due to the significant societal consequences caused by the Laki eruption 1783–1784 CE (15 km^3 lava, estimated 120 Tg SO_2, Thordarson and Self, 2003) across the northern hemisphere. During the Laki eruption, a thick sulphate aerosol veil and fluoride contamination of pasture and drinking water for livestock, led to the loss of ∼20% of Iceland's population through famine (Thordarson and Self, 2003). It has been suggested that mortality in the UK and mainland Europe also increased (Schmidt et al., 2011; Witham and Oppenheimer, 2004). Large fissure eruptions are often assumed to be the closest analogues for large igneous province volcanism during the human era (e.g., Self et al., 2014) and so understanding them is further important in order to gain deeper insights into these massive environmental perturbations that punctuate Earth history. The Holuhraun eruption in Iceland in 2014–2015 (Fig. 1) is the largest such event since Laki and presents us with a rare opportunity to study volcanism of this style and scale using modern scientific techniques.

Tropospheric plumes from eruptions with low-level explosivity are complex mixtures of magmatic gases, entrained background air and aerosol particulate matter (PM), and usually low quantities of silicate ash. Volcanic plumes are known to contain nearly all naturally occurring elements (e.g., Allard et al., 2000; Symonds et al., 1992) ranging from the common metals (Na, K, Ca, Mg) to trace species (e.g., Pb, Mo, As, Se, Hg and Br). The interactions of this range of elements with ecosystems are highly variable and deposition of plumes can be either adverse or beneficial for terrestrial and aquatic environments (e.g. Cronin and Sharp, 2002; Jones and Gislason, 2008).

Volcanic gases and PM may also present a health hazard through air pollution. Sulphur dioxide (SO_2) levels in particular often exceed official air quality standards for this gas in the ambient air (Gíslason et al., 2015; Schmidt et al., 2015; Tam et al., 2016). A large proportion of volcanic PM is in the $PM_{2.5}$ fraction (sub-2.5 μm diameter), which has become a key metric in air pollution guidelines. Short and long term exposure to $PM_{2.5}$ in the ambient air is estimated to cause over 3 million premature deaths globally per year (Lim et al., 2012), and it remains the single largest environmental health risk in Europe (Holgate, 2017). Detailed information on SO_2 and PM size-resolved mass flux, and gas and PM composition, is therefore necessary for reliable assessments of the effect of volcanic emissions on the environment and health.

The Holuhraun eruption occurred in the central highlands of Iceland (Fig. 1), on the ice-free part of Bárðarbunga volcanic system and lasted 6 months from 31 August 2014 to 28 February 2015. The estimates of its total SO_2 emission range between 11 Tg (Gíslason et al., 2015) and 19 Tg (Gauthier et al., 2016), 1.5–2 times more than the total anthropogenic SO_2 emissions from the European Economic Area in 2011 (European Environmental Agency, 2017). Daily SO_2 gas fluxes averaged 60 kt/day (Gíslason et al., 2015), a rate of outgassing rarely observed during sustained eruptions. It dwarfed most other long-term volcanic SO_2 emitters, such as Kilauea volcano in Hawaii (2–8 kt/d, Elias and Sutton, 2012) and Mt Etna, Italy (3.5 kt/d between 2005 and 2008, Salerno et al., 2009). It was one of the largest global tropospheric volcanic emissions since Laki, rivalled only by emission from Miyakejima 2000–2003 (∼18 Mt SO_2, Kazahaya et al., 2004). This caused frequent and persistent deterioration of air quality in Iceland with regard to SO_2 (Gíslason et al., 2015), and an impact on the chemistry of snow and rain precipitation around Iceland (Galeczka et al., 2016; Gíslason et al., 2015; Stefánsson et al., 2017). Long-range periodic impacts on air quality were reported in the UK, Ireland, Finland, the Netherlands and Austria (Gíslason et al., 2015; Schmidt et al., 2015; Twigg et al., 2016).

Our study quantifies the impact of Holuhraun on populated areas in Iceland. Specifically we trace the evolution of the plume chemistry from the eruption site to 2 key areas of population: Reykjahlíð, which is the nearest municipality to Holuhraun at 100 km distance, and Reykjavík capital area, which hosts ∼60% of Iceland's population, 250 km distance (Fig. 1). We present a novel dataset of gas and size-resolved aerosol chemistry, including a time series of constituents relevant for air quality assessment, i.e., SO_2, $PM_{2.5}$ and sulphate aerosol, and analysis of trace species including heavy metals. The findings of this study are relevant for multiple other countries impacted by volcanic emissions, either through prolonged eruptions, for example that of Kilauea in Hawaii, or through persistent quiescent degassing, such as from Masaya in Nicaragua.

2. Methodology

Data were collected at 3 sites (Fig. 1, section 2.1). The instrumentation and analytical techniques are detailed in sections 2.2–2.6.

2.1. Site descriptions

2.1.1. Holuhraun eruption site

The field campaign took place 21–22 January 2015, when we had favourable winter weather, dry and generally bright, thick

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