

Introduction

Iceland is one of the most active volcanic regions on Earth, with more than 30 active volcanic systems erupting about 20 times, and producing more than 5 km³ magma per century (Thordarson and Hoskuldsson, 2008). These eruptions can have significant effects upon the natural and human environment. Many of the volcanoes are covered with glaciers and often generate volcanic ash and occasionally, “Amazonian” sized floods (~200,000 m³ s⁻¹). Because of Iceland’s location and prevailing winds, volcanic ash and gases can pollute the atmosphere and disturb air traffic over Western Europe (EC, 2010). The ash fallout and floods can both fertilise and pollute surface waters (Flaathen and Gislason, 2007).

The 2014–2015 eruption within the Bárðarbunga volcanic system began in the morning of 31 August 2014 following a minor eruption at the same site (<0.001 km³) that lasted only a few hours, two days earlier. The eruption ended on 27 February 2015 after six months of continuous activity, generated minute amounts of volcanic ash, large emissions of gases, and a basaltic lava flow referred to here as the Holuhraun lava (the largest by volume in Iceland since the 1783–1784 Laki eruption).

The Laki eruption produced in eight months about ten times more lava and SO₂ (120 Mt) than Holuhraun (Thordarson and Self, 2003). The environmental pressure of the Laki eruption was enormous: in Iceland about 60 % of the grazing livestock and 20 % of the human population died (Thordarson and Self, 2003). The average monthly emission rates were greater than the peak global anthropogenic SO₂ emission rate in 1970–1980 (133 Mt yr⁻¹) and twice that from the combined USA, Canada, Western and Central Europe at the peak emissions in the 1970s (76 Mt yr⁻¹; Smith *et al.*, 2011). These peak man-made emissions caused severe environmental impacts including acid rain and the mobilisation of aluminium from rocks and soil, degrading aquatic and terrestrial ecosystems (Gensemer and Playle, 1999). Since the peak, all these emissions have been declining and were down to 14.7 Mt yr⁻¹ from USA, Canada, Western and Central Europe in 2011 (Klimont *et al.*, 2013). This decline is confirmed by long term precipitation and air quality monitoring in South Iceland, with higher annual pH and decreasing variability, and fewer extreme incidents, in years without volcanic eruptions (Sigurdsson and Thorlacius, 2014). Furthermore, the average dissolved river water sulphur concentration was reduced by 40 % from 1970 to 2004 (Gislason and Torssander, 2006).

The 2014–2015 Bárðarbunga eruption provides an opportunity to study the environmental pressure from a relatively large emission source. Direct gas measurements and petrologic gas estimates along with detailed field observations and modelling allow this eruption to be compared with large historical eruptions that caused major environmental problems. Here we report on the effusion rate, gas emissions, and the environmental pressure from this eruption.

Environmental pressure from the 2014–15 eruption of Bárðarbunga volcano, Iceland

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Abstract

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The effusive six months long 2014–2015 Bárðarbunga eruption (31 August–27 February) was the largest in Iceland for more than 200 years, producing 1.6 ± 0.3 km³ of lava. The total SO₂ emission was 11 ± 5 Mt, more than the amount emitted from Europe in 2011. The ground level concentration of SO₂ exceeded the 350 µg m⁻³ hourly average health limit over much of Iceland for days to weeks. Anomalously high SO₂ concentrations were also measured at several locations in Europe in September. The lowest pH of fresh snowmelt at the eruption site was 3.3, and 3.2 in precipitation 105 km away from the source. Elevated dissolved H₂SO₄, HCl, HF, and metal concentrations were measured in snow and precipitation. Environmental pressures from the eruption and impacts on populated areas were reduced by its remoteness, timing, and the weather. The anticipated primary environmental pressure is on the surface waters, soils, and vegetation of Iceland.

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Results

Methods are described in the Supplementary Information and include: location of the monitoring stations in Iceland (Fig. S-1), gas emission measurements (Table S-1), ground-level SO₂ gas concentrations in Europe (Table S-2), SO₂ dispersion during the eruption including the effect of precipitation (Fig. S-2) and wind-speed distribution over the year at the eruption site (Fig. S-3).

The lava and effusion rate. Apart from modest fire fountaining in the first few weeks, the volcanic activity was effusive, forming a lava flow that advanced 17 km towards NNE in 15 days. As time progressed, further lobes of lava developed and grew rapidly in surface area. During December and January thickening by inflation was prominent with a much slower areal growth rate (Fig. S-1). At the end of the eruption, the lava covered an area of $84.1 \pm 0.6 \text{ km}^2$ and its volume was $1.6 \pm 0.3 \text{ km}^3$. Using a bulk lava density of 2600 kg m^{-3} , the total mass erupted was $(4.2 \pm 0.8) \times 10^{12} \text{ kg}$. The average magma flow rate for the 181 days of eruption was $100 \text{ m}^3 \text{ s}^{-1}$ and mass flow rate $2.7 \times 10^5 \text{ kg s}^{-1}$. During the first weeks, the flow rate was two to three times the average, while flow rates of $50\text{--}80 \text{ m}^3 \text{ s}^{-1}$ occurred in December and January followed by gradual decline in February leading to the termination on 27 February 2015.

The erupted lava is olivine tholeiite and its composition was constant during the eruption. The rock is poor in macrocrystals, and is mainly plagioclase but olivine and clinopyroxene occur in minor amounts. The groundmass is composed of silicate glass and microcrystals of plagioclase, clinopyroxene, and olivine. The sulphur-rich nature of the lava is reflected by Fe-rich sulphide globules in the groundmass. The H₂O content of melt inclusions is as high as 0.5 wt. % whereas CO₂ reaches 900 ppm. Groundmass glass contains 0.1 wt. % H₂O and no CO₂. The pure CO₂ inclusions, associated with melt inclusions, indicate that the basalt was CO₂ saturated when the inclusions were trapped from partially degassed basalt before, and deeper than, the onset of H₂O and S degassing. The CO₂ saturation occurred at approximately 3 kbar pressure (~9 km depth).

Petrologic estimates of gas emission. Taking into account the average extrusion rate, the total H₂O and CO₂ outputs were 16.8 ± 3.6 and $6.33 \pm 1.7 \text{ Mt}$ respectively, giving average emission rates of 1074 ± 231 and $405 \pm 106 \text{ kg s}^{-1}$. The integrated SO₂ emission was $10.7 \pm 3.0 \text{ Mt}$ and average emission rate was $684 \pm 193 \text{ kg s}^{-1}$. Chlorine degassing was uncertain but a maximum atmospheric load was estimated as 0.1 Mt HCl, yielding an average flux of 6.4 kg s^{-1} . No HF degassing can be inferred from the lava. All of these fluxes changed over time as shown in Table S-1 in the Supplementary Information.

Direct measurements of gas emissions. The emission rate of SO₂ was monitored from the beginning of the eruption with ground-based DOAS (Differential Optical Absorption Spectroscopy) and the ratio of SO₂ to CO₂, H₂S, H₂ and H₂O were measured with MultiGAS. Ten complete DOAS traverses were made 80–240 km from the vent (Fig. S-1) between 21 September and 4 February. These show a temporal decrease in SO₂ emissions $1400\text{--}100 \text{ kg s}^{-1}$. Applying a

linear fit to the data ($r^2 = 0.76$), yields $11.8 \pm 4.1 \text{ Mt}$ of SO₂ emission during the eruption. Eleven MultiGAS acquisitions of sufficient quality were made during 30 August–21 January. The eleven CO₂/SO₂ ratios and five H₂O/SO₂ ratios decrease over time. The measured eruption plume became enriched in SO₂ relative to CO₂ and H₂O, even though the total emission of SO₂ decreased. This reflects changes at the vent, less glacier melting with time, and the increasing distance from vent to sampling stations as the lava field extended (Fig. S-1). Knowing that $11.8 \pm 4.1 \text{ Mt}$ of SO₂ were emitted, and the MultiGAS ratios, the total emissions of CO₂ and H₂O were 5.6 ± 3.6 and $284 \pm 193 \text{ Mt}$ respectively. The emissions are not exclusively magmatic in origin, particularly in the beginning of the eruption when there was a large contribution from meteoric water.

Air quality simulations and measurements. Variable wind conditions have played a major role in gas distribution over Iceland. The CALPUFF air quality model, initialised with emission rates estimated from the DOAS measurements, has been used to simulate the dispersal of the SO₂ cloud and its surface concentration. The results are presented in Figure 1 as the frequency of hourly concentrations higher than the $350 \mu\text{g m}^{-3}$ limit (Icelandic Health Regulation, 2002). The values corresponding to each contour show how many times this

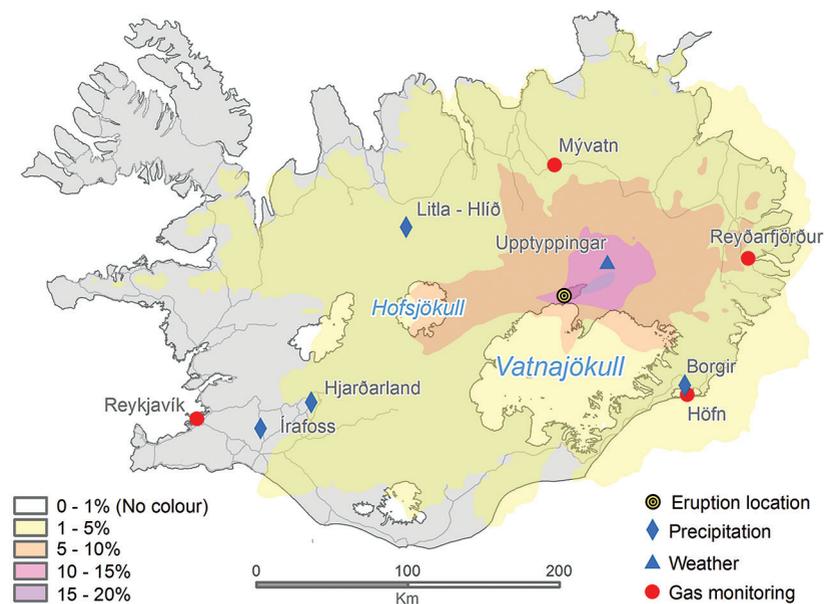


Figure 1 The modelled SO₂ dispersion during the eruption, presented as the frequency of hourly concentrations higher than the $350 \mu\text{g m}^{-3}$ health limit. The monitoring stations mentioned in the text are also shown.



concentration has been exceeded at each location during this period. Gas pollution has been extensive across all of Iceland. The NE part of the country has suffered the highest impact from the eruption. The model suggests that an area 50 km NE of the eruption site exceeded $350 \mu\text{g m}^{-3}$ for up to 20 % of the time (about 30 days in total). The northern part of Vatnajökull and the eastern part of Hofsjökull glaciers have frequently been exposed to high ground-level concentrations of SO_2 for up to 15 days.

Prior to the Holuhraun eruption, the ground-level concentration of atmospheric SO_2 in Iceland had never been recorded as exceeding the $350 \mu\text{g m}^{-3}$ hourly limit. The first permanent monitoring station was installed in 1968. During the eruption, predicted and measured values repeatedly surpassed this limit (Figs. 1 and 2). Much higher instantaneous SO_2 peaks were frequently measured on all mobile stations (Fig. S-1), the highest being $21,000 \mu\text{g m}^{-3}$ in Höfn. Continuous measurements started 28 October 2014 in Höfn as shown in Figure 2. There the hourly averaged concentration reached a maximum of $3050 \mu\text{g m}^{-3}$ on 11 January 2015. Over the monitoring periods shown in Figure 2, SO_2 exceeded

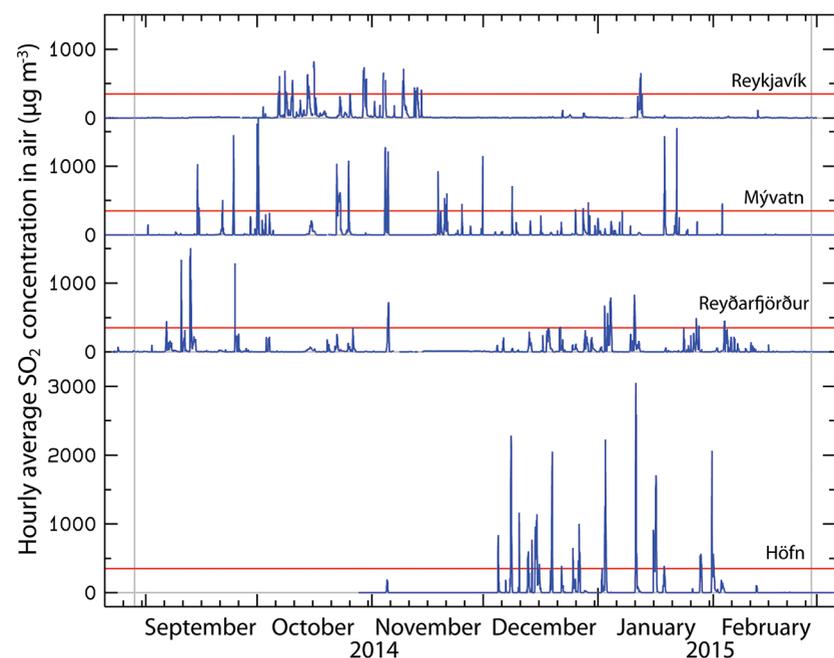


Figure 2 The SO_2 concentration in air at four of the permanent gas monitoring stations presented in Figure 1. The $350 \mu\text{g m}^{-3}$ health limit is shown by the red horizontal line. The grey vertical lines mark the eruption period. Permanent SO_2 monitoring started at Höfn 28 October 2014.

the $350 \mu\text{g m}^{-3}$ threshold 2.0 % of the time at Mývatn (for 17 consecutive hours and a total of 86 hours), 1.4 % in Reyðarfjörður (for 10 consecutive hours and a total of 58 hours), 1.4 % in Reykjavík (for 8 consecutive hours and a total of 59 hours) and 4.2 % of the time in Höfn (for 16 consecutive hours and a total of 124 hours). The last unambiguous detection of the volcanic plume was at the Mývatn station on February 18. Gas emissions from Holuhraun increased ground-level SO_2 concentrations in Europe in September 2014 (Schmidt *et al.*, 2015, Tables S-1 and S-2), when the eruption intensity was high.

Dissolved constituents in precipitation. During the eruption, 648 precipitation samples were collected from 21 stations (Fig. S-1). Low pH values were mainly observed in Southern Iceland, the lowest was 3.2, at Borgir near Höfn (Figs. 3, 1 and S-1). At Árafoss, a long term air quality and precipitation monitoring station in the SW, the average pH was 5.4 (4.0–6.6) with 25 % of the samples having a pH lower than 5, during the eruption. This value is lower than that recorded in years not influenced by volcanic eruptions (2005–2009) when the pH was 5.8 (4.5–7.7). Many of the pH measurements done during the eruption were high due to dust-water interactions as detailed in the Supplementary Information. At Árafoss, during the eruption, the average concentration of SO_4 was $426 \mu\text{M}$ (range 1.8–12,270) and F $17.7 \mu\text{M}$ (range <0.2–444). These values are 39 and 68 times greater than those recorded in previous years ($11 \mu\text{M}$, range 2.5–27) and ($0.26 \mu\text{M}$, range 0.01–1.7) for SO_4 and F respectively. The stations with the most elevated SO_4 and F concentrations were Litla-Hlíð and Hjarðarland (Fig. 1).

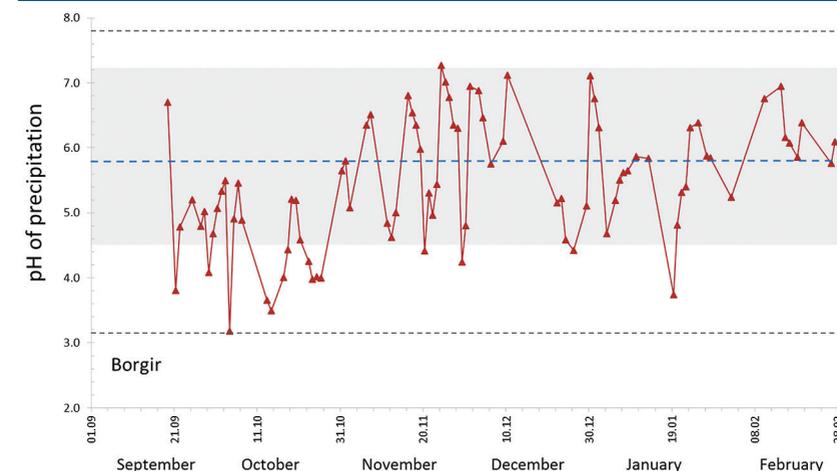


Figure 3 The pH of precipitation at Borgir during the eruption (Figs. 1 and S-1). The blue dashed line represents the average pH (5.8) and the shaded background the pH range for the eruption free period 2005–2009 at the Árafoss monitoring station (Figs. 1 and S-1). The two grey dashed lines depict the variation in pH during the eruption from the monitoring stations shown in Figure S-1.



The conductivity and pH of the melted snow collected in the vicinity of the Holuhraun lava ranged over 5–234 $\mu\text{S cm}^{-1}$ and 3.34–4.85 respectively. The snow was acidified by H_2SO_4 , HCl , and HF (Fig. 4). About half of the anions were balanced by protons (H^+) and the rest by cations such as Al^{3+} , Fe^{3+} , Fe^{2+} , Na^+ , Ca^{2+} , and Mg^{2+} derived from water soluble metal salts. The overall sea spray concentration addition was less than 5.15 μM for Cl. The concentrations of F, Al, Fe, Mn, Cd, Cu, and Pb in the snow surpassed drinking-water standards (European Community, 1998).

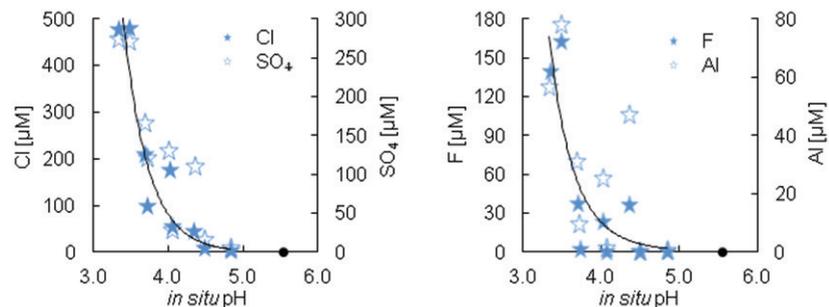


Figure 4 The Cl, SO_4 , F, and Al concentration in melted snow (Fig. S-1) versus *in situ* pH (0.1 °C). The black dot represents the pH (5.54) of pure water equilibrated with 395 ppmv of CO_2 at 0.1 °C.

Discussion

The eruption intensity declined continuously from start to finish (Table S-1). In that period gas pollution affected most of Iceland (Fig. 1) and occasionally reached Europe (Table S-2; Schmidt *et al.*, 2015). Although Northern Iceland more frequently experienced high SO_2 concentrations, the lowest pH of precipitation was measured in the south. This could be due to differences in exposure time to light and humidity between relatively the high and low latitude travel paths of the plume, resulting in discrepancies in the oxidation rate of SO_2 and rain acidification (Lelieveld 1993; Schmidt *et al.*, 2010). Furthermore suspension of dust, and the resulting water-particle interactions could raise pH (Gislason and Eugster 1987), reducing the solubility of most polluting metals. Iceland is one of the most active dust sources on Earth with dust flux of 30–40 Mt per year (Arnalds *et al.*, 2014) mainly coming from the glacier outwash planes.

The concentration of dissolved metal salts in the melted snow was equal to that of the acids. These metals are soluble at low pH and can both pollute and fertilise surface waters. For example, the highest Al concentration in Figure 4 is about 5 times greater than the Al concentration that decreases the life expectancy of salmon fry by 50 % (Gensemer and Playle, 1999). The frequent low pressure

systems passing over Iceland provided strong winds (Fig. S-3), fast dispersion of the eruption plume, dust suspension, and thaw periods, preventing build-up of pollutants in the snow pack at low altitude. However, pollutants may have accumulated in the snow at high altitude. The first fraction of meltwater in the spring can contain more than five times the average snowpack concentration of pollutants, and may enter streams and lakes without contact with soil (Johannessen and Hendriksen, 1978).

Petrologic estimates and direct measurements of SO_2 and CO_2 emission from Holuhraun are in agreement. This lends credence to the petrologic emission rates estimated for historical eruptions (*e.g.*, Thordarson and Self, 2003). Based on this evidence, the Holuhraun emissions were an order of magnitude lower than the Laki emissions that started 8 June 1783. The average wind speed is higher in winter than summer (Fig. S-3), thus the Holuhraun eruption produced faster-dispersing plumes. Because of reduced autumn-winter sunlight hours, a smaller per cent of emitted SO_2 had the potential to be oxidised under dry conditions to H_2SO_4 in the Holuhraun eruption compared with the Laki eruption (Schmidt *et al.*, 2010). During winter there is perhaps greater environmental and human health risk from SO_2 than from sulphate aerosol particles due to reduced conversion efficiency, whereas in summer the aerosol particle effects may dominate.

Conclusions

The location and timing of the Holuhraun eruption in many ways minimised its impact on humans and the environment. It was located: 1) far away from inhabited parts of the island, 2) just north of the Vatnajökull glacier edge which narrowly escaped volcanic ash deposition and ensuing floods, 3) in the glacier's rain shadow leading to relatively low H_2SO_4 formation since oxidation of SO_2 is slow in dry air compared to humid (Lelieveld, 1993), 4) in an abundant, acid-neutralising dust source. The eruption occurred at a time when: 5) vegetation growth was at its minimum, 6) grazing animals were moving towards lower altitudes, 7) SO_2 oxidation in dry air was minimised due to limited sunlight in autumn-winter, and 8) average wind speeds were high, dispersing the plume and mobilising dust.

Because of these factors, the environmental pressure of the Holuhraun eruption is expected to be moderate, while large enough to be measurable because the rate of SO_2 emitted from this point source was considerably greater than current anthropogenic emission rate in Europe.

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Additional Information

Supplementary Information accompanies this letter at www.geochemicalperspectivesletters.org/article1509

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