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## Balloon-borne measurement of the aerosol size distribution from an Icelandic flood basalt eruption



D. Vignelles <sup>a,\*</sup>, T.J. Roberts <sup>a</sup>, E. Carboni <sup>b</sup>, E. Ilyinskaya <sup>c</sup>, M. Pfeffer <sup>d</sup>, P. Dagsson Waldhauserova <sup>e,f,g</sup>, A. Schmidt <sup>c</sup>, G. Berthet <sup>a</sup>, F. Jegou <sup>a</sup>, J.-B. Renard <sup>a</sup>, H. Ólafsson <sup>h</sup>, B. Bergsson <sup>d</sup>, R. Yeo <sup>d</sup>, N. Fannar Reynisson <sup>d</sup>, R.G. Grainger <sup>b</sup>, B. Galle <sup>i</sup>, V. Conde <sup>i</sup>, S. Arellano <sup>i</sup>, T. Lurton <sup>a</sup>, B. Coute <sup>a</sup>, Vincent Duverger <sup>a</sup>

- <sup>a</sup> LPC2E/CNRS, Université d'Orléans, 3A, Avenue de la Recherche Scientifique, 45071 Orléans, France
- <sup>b</sup> COMET, Atmospheric, Oceanic and Planetary Physics, University of Oxford, Parks Road, Oxford, OX1 3PU, UK
- <sup>c</sup> School of Earth and Environment, University of Leeds, LS2 9JT, Leeds, UK
- <sup>d</sup> Icelandic Meteorological Office, Bústaðavegi 7–9, 150 Reykjavik, Iceland
- <sup>e</sup> University of Iceland, Departments of Physical and Earth Sciences, Reykjavik, Iceland
- <sup>f</sup> Agricultural University of Iceland, Faculty of Environment, Hvanneyri, Iceland
- <sup>g</sup> Czech University of Life Sciences Prague, Prague, Czech Republic
- h University of Iceland, Icelandic Meteorological Office and Institute for Meteorological Research, Reykjavík, Iceland
- <sup>1</sup> Chalmers University of Technology, Department of Earth and Space Sciences, Hörsalsvägen 11, 412 96 Gothenburg, Sweden

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#### ABSTRACT

We present in situ balloon-borne measurements of aerosols in a volcanic plume made during the Holuhraun eruption (Iceland) in January 2015. The balloon flight intercepted a young plume at 8 km distance downwind from the crater, where the plume is  $\sim$ 15 min of age. The balloon carried a novel miniature optical particle counter LOAC (Light Optical Aerosol Counter) which measures particle number concentration and size distribution in the plume, alongside a meteorological payload. We discuss the possibility of calculating particle flux by combining LOAC data with measurements of sulfur dioxide flux by ground-based UV spectrometer (DOAS).

The balloon passed through the plume at altitude range of 2.0–3.1 km above sea level (a.s.l.). The plume top height was determined as 2.7–3.1 km a.s.l., which is in good agreement with data from Infrared Atmospheric Sounding Interferometer (IASI) satellite. Two distinct plume layers were detected, a non-condensed lower layer (300 m thickness) and a condensed upper layer (800 m thickness). The lower layer was characterized by a lognormal size distribution of fine particles (0.2  $\mu$ m diameter) and a secondary, coarser mode (2.3  $\mu$ m diameter), with a total particle number concentration of around 100 cm<sup>-3</sup> in the 0.2–100  $\mu$ m detection range. The upper layer was dominated by particle centered on 20  $\mu$ m in diameter as well as containing a finer mode (2  $\mu$ m diameter). The total particle number concentration in the upper plume layer was an order of magnitude higher than in the lower layer.

We demonstrate that intercepting a volcanic plume with a meteorological balloon carrying LOAC is an efficient method to characterize volcanic aerosol properties. During future volcanic eruptions, balloon-borne measurements could be carried out easily and rapidly over a large spatial area in order to better characterize the evolution of the particle size distribution and particle number concentrations in a volcanic plume.

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#### 1. Introduction

Volcanoes release gases and particles into the atmosphere through continuous degassing or episodic eruptive events, and depending on the injection altitude and emission rate, they can im-

\* Corresponding author.

E-mail address: damien.vignelles@cnrs-orleans.fr (D. Vignelles).

pact both the tropospheric and stratospheric composition and climate (McCormick et al., 1995; Robock, 2000; Schmidt et al., 2012; Solomon et al., 2011). Ash-rich plumes such as that from the Icelandic Eyjafjallajökull eruption in 2010 can lead to widespread disruption of aviation (Spinetti et al., 2013). Ash-poor volcanic plumes may also strongly impact the environment and quality of life due to high concentrations of polluting gases and aerosol

particles. Indeed, the recent flood basalt eruption at Holuhraun (31 August 2014–27 February 2015, 1.6 km<sup>3</sup> of erupted lava, Gislason et al., 2015) was a major source of sulfur gases and aerosols and caused both local (Gislason et al., 2015) and European-wide (Schmidt et al., 2015) deteriorations to air quality.

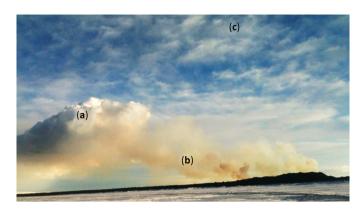
Long-lasting flood basalt eruptions are one of the most hazardous volcanic scenarios in Iceland and have had enormous societal and economic consequences across the northern hemisphere (Gudmundsson et al., 2008). One of the best known examples is the Laki eruption (1783-84 CE) (Thordarson and Self, 2003) which led to deaths of >20% of the Icelandic population by environmental pollution and famine, and likely increased European levels of mortality through air pollution by sulfur-bearing gas and aerosol (Grattan, 1998; Taylor et al., 2003; Witham and Oppenheimer, 2004). Potential impacts of such an eruption on modern day Europe have been modeled by Schmidt et al. (2011) who found that PM2.5 aerosol pollution would double causing 142,000 additional cardiopulmonary fatalities in the year following the eruption onset. A Laki-type eruption scenario has been recently included in the UK National Risk Register (UK Cabinet Office, 2013). However, there are still many uncertainties about the source terms of Icelandic flood basalt eruptions that are necessary for atmospheric models and health impact assessments. The 2014-2015 Holuhraun eruption was therefore a unique opportunity to study the nearsource composition of an Icelandic flood basalt eruption plume.

Direct measurements of volcanic aerosol (defined here as nonsilicate particles such as sulfate) are needed to better constrain the plume sulfur chemistry and particle processes, which together with plume injection height are two key uncertainties in models used to predict the dispersion and air quality impacts from eruptions. Existing in-situ measurements of elevated volcanic plumes mostly involve interception of aged plumes that have already undergone significant chemical and physical evolutions (Marenco et al., 2011; Jégou et al., 2013). Small portable sensors placed on airborne drones or balloons offer new possibilities to characterize volcanic plumes close to source. McGonigle et al. (2008) demonstrated heli-type drone sensing of SO<sub>2</sub> and CO<sub>2</sub> to determine CO<sub>2</sub> fluxes at Vulcano fumarole field (Italy). More recently, Shinohara (2013) deployed a suite of gas sensors on a drone to characterize the plume of Kirishima volcano (Japan) during an eruptive phase where ground-based sampling was too hazardous. Pieri et al. (2013) performed drone as well as balloon-based campaigns to measure gases and ash particles in the eruptive plume of Turrialba volcano (Costa Rica).

Here we present measurements made by a newly developed lightweight optical aerosol counter (LOAC) carried on a meteorological balloon through the near-source Holuhraun eruption plume. By combining size-resolved particle number concentration measurements with meteorological parameters and remote sensing of  $SO_2$  flux, we are able to provide some of the key eruption source term information.

#### 2. Holuhraun and plume conditions

Holuhraun is located northwards of the Vatnajökull ice cap in the largest desert area of Iceland. On January 22nd 2015, visible plumes were emitted from the main crater (Baugur) and several places within the lava field (Fig. 1). It was estimated that ~90% of the released gas volume was from Baugur. These distinct plumes merged into one main plume which was advected northeastwards. The rising plume visibly changed while being advected, with the upper part turning into a condensed, optically thick cloud several kilometers downwind of the source. The atmosphere was very clear within the boundary layer, and the lower troposphere was cloud-free except for the volcanic plume. Clouds were visible at a much higher altitude (>5.5 km above the sea level (a.s.l.) as



**Fig. 1.** Picture taken at 14 UTC on January 22nd 2015 during the afternoon before the balloon flight. a) Condensed plume, b) non-condensed plume, c) high altitude clouds.

described in Section 4). Fig. 1 shows the conditions at the eruption site on January 22nd at 14 UTC. The visual appearance of the plume remained consistent throughout the day. Plume dispersion modeling (Iceland Met Office, CALPUFF model, Barsotti et al., 2008) predicted that the northeastward plume advection continued during the night of January 22nd when our balloon-borne measurements were made (see online Appendix 1 for further details). Although the CALPUFF model predicts only ground-level plume exposure, the constant vertical wind profile calculated by HARMONIE model for Holuhraun area (see Section 4) allows us to assume a consistent plume transport direction from ground level up to 4 km a.s.l.

#### 3. Methods

#### 3.1. Balloon instrumentation

The LOAC (Light Optical Aerosol Counter) is an optical particle counter sufficiently lightweight to be carried by a 1000 g meteorological balloon. The instrument contains a laser (650 nm) and measures the intensity of light scattered at two angles, 12° and 60° (Lurton et al., 2014; Renard et al., 2016) to discriminate the particle concentration over 19 size classes from 0.2 µm to 100 µm in diameter. Sampling is driven by a miniature pump (constant flowrate of 2 Lmin<sup>-1</sup>) enclosed in the gondola with the air pumped through the measurement cell and released afterwards. For the LOAC integration time of 10 s, the counting uncertainty is derived from the Poisson counting statistics and defined as one relative standard deviation: 60% for a particle number concentration of  $10^{-2}$  cm<sup>-3</sup>, 20% for  $10^{-1}$  cm<sup>-3</sup> and 6% for a particle number concentrations higher than 1 cm<sup>-3</sup>. A complete description of the instrument can be found in Renard et al. (2016). Differences in scattering between the two distinct angles are also used to determine the typology. The typology is a specific term related to the main refractive index of particles sampled, obtained by combining the intensities of light scattered at two diffusion angles (Renard et al., 2016). This can provide information on the nature of the particles, determined by reference to laboratory measurements (Renard et al., 2016). The typology gives several classes of optical properties discussed in Section 5.3. The aerosol data, GPS coordinates, temperature and hygrometry are sent in real time by a telemetry system. The flight chain configuration is shown in Fig. 2.

To aid interpretation, the balloon data are analyzed in conjunction with model outputs (backward trajectories, air quality plume dispersion and meteorological model) and remote sensing data (ground-based DOAS, and satellite Infrared Atmospheric Sounding Interferometer (IASI) overpasses) in the Results Section 4.

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