

SM1: Size-resolved chemistry of the PM

Size-resolved chemistry of PM collected using cascade impactors at (i) a near-vent location (~5km from eruptive source), and (ii) a downwind location (~100km downwind, Reykjavíð town). Major ions (Cl^- , SO_4^{2-} , Br^-): The dominant size bin in the near-vent PM is 0.1-0.18 μm diameter (sulphate peak is spread between 0.1-0.56 μm). Sulphate has a significant secondary peak on the inlet stage, in PM >18 μm . The halogens (Cl^- and Br^-) are also abundant in the finest size bin resolved by the impactor (0.032-0.018 μm) and the after-stage which collects all PM <0.018 μm (plotted on the graph as 0.001 μm). In addition, Cl^- has a secondary peak in 5.6-3.2 μm size bin. Sulphate in the downwind sample peaks at 0.18-0.56 μm , which is very similar to the near-vent samples, albeit a little coarser. Size bins below 0.18 μm are not resolved by the impactor used so we cannot directly compare the 0.1-0.18 μm bin with the near-vent sample. However, the after-stage (shown on the plot as 0.001 μm) captures all PM <0.18 μm so if there was significant amount of PM in the smaller size bins it would be seen on the after-stage. This appears not to be the case. The halogens (Cl^- and Br^-) peak in the larger size bin 0.56-5.6 μm . Sulphate is not nearly as abundant relative to Cl^- in the downwind sample as in the near-vent sample.

Trace elements: In the near-vent PM most elements have their concentration peak in the size bin 0.1-0.18 μm . A number of elements also have elevated concentrations in even finer size bins, including the after-stage which is all PM <0.018 μm (shown on the plots as 0.001 μm). The size distribution is not absolutely clear-cut for all elements, and a number of them appear to have no preference for a particular size bin. Size distributions in the downwind PM is much more spread between the size bins (FigIM-TE). The dominant size bin is 0.18-0.32 μm but there are also peaks in 1.0-1.8, 3.2-5.6 and 10-18 μm size fractions.

Figure SM1. Size-resolved chemical composition of near-vent and downwind PM

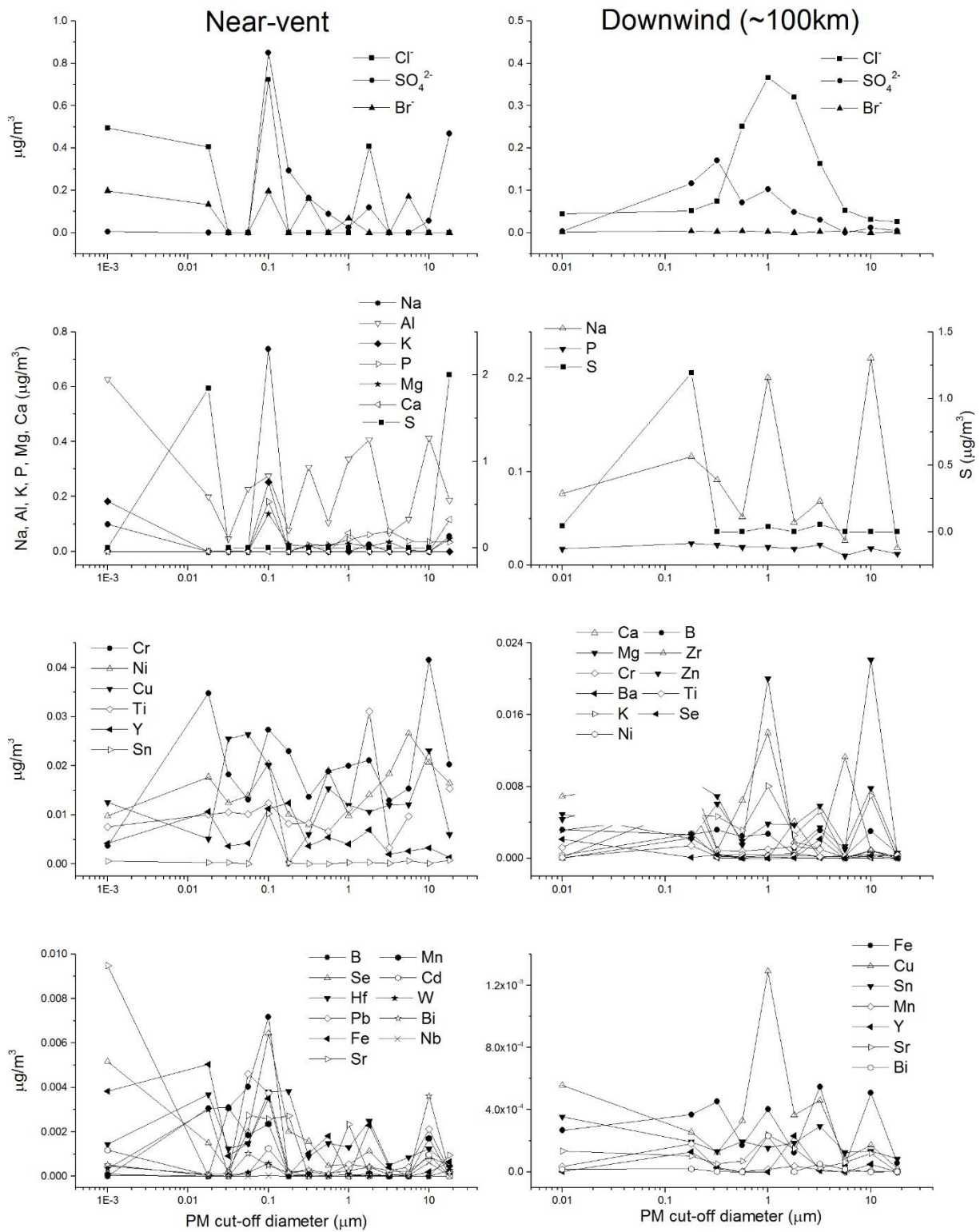
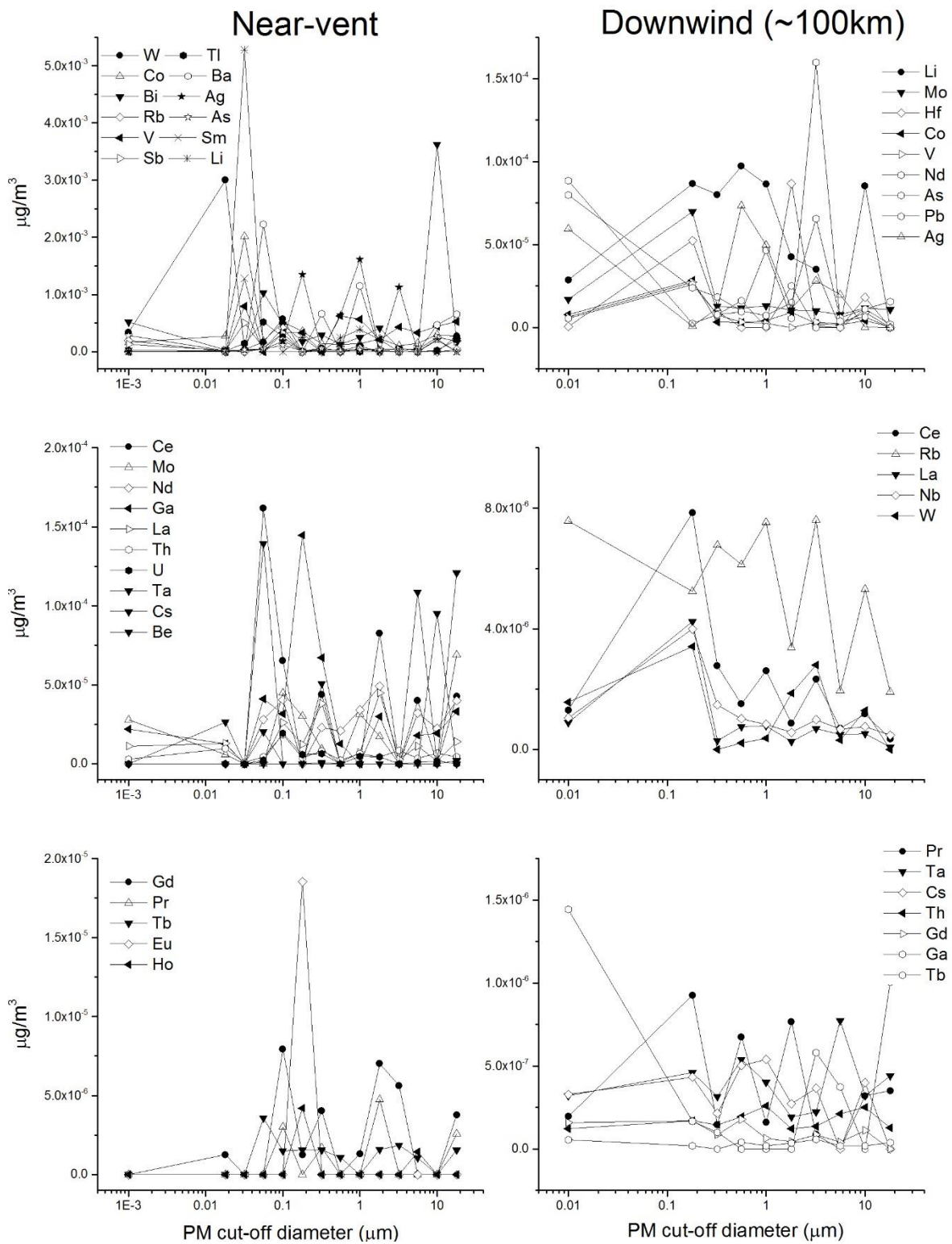


Figure SM1 continued



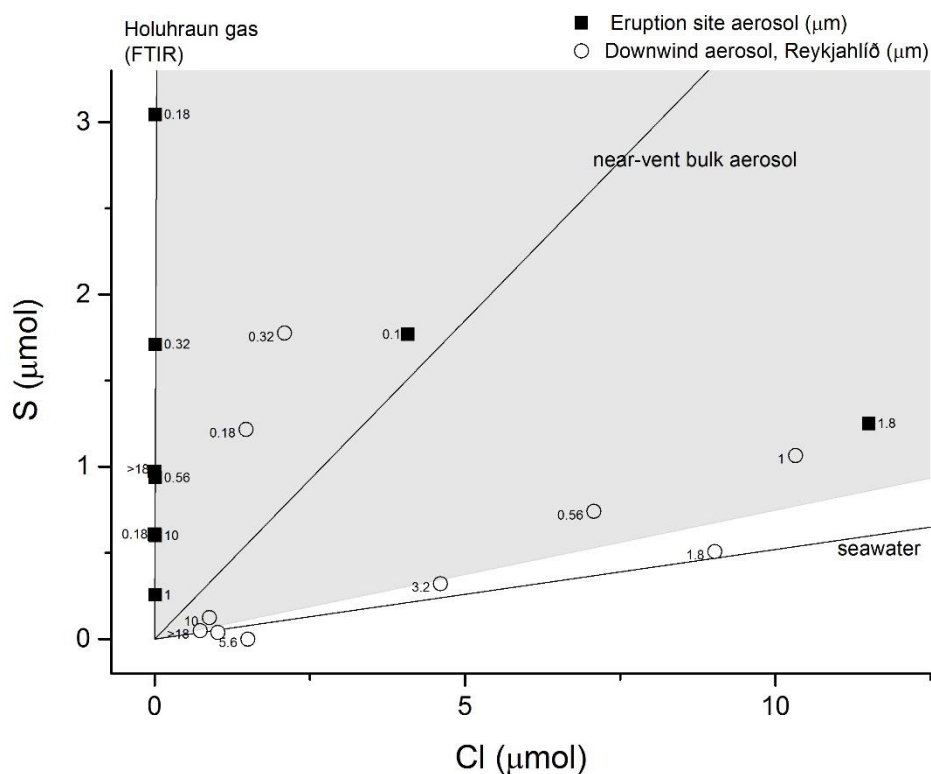
SM2: Source appointment of PM in volcanic plume at different distance from the emission source

Figure SM3 shows molar S/Cl ratio in PM sampled in two locations, near-vent (~5km from emission source) and downwind (~100 km from emission source). The plot also shows S/Cl in Holuhraun magmatic gas (Burton et al. in review), bulk PM collected near-vent by filter packs (this study), and seawater (S/Cl = 0.05, Bruland, 1983) which is a dominant source of background aerosol in Iceland (e.g. Stefánsson et al., 2017). While wind-blown rock dust is another important source of aerosol in Iceland, it does not impact the concentration of S and Cl anions (Stefánsson et al., 2017 and references within).

The bulk PM data is shown as the mean and standard deviation of 5 samples (S/Cl = 0.37 ± 0.30 , grey line and shaded area). For the size-resolved impactor samples, the cut-off diameter of the PM is shown next to each data point. Seawater S/Cl ratio is much lower than the 'plume' signature in gas and PM, although the PM ratio is very variable.

The near-vent size-resolved samples appear to have a volcanic source signature with a high S/Cl ratio for most size bins, falling very close to the gas phase ratio. The exceptions are the size bins 0.1-0.18 and 1.8-3.2 μm which have a lower S/Cl ratio but still fall within the 'bulk aerosol' shaded area. This strongly suggests that near-vent aerosol in all of the resolved size bins are dominated by a volcanic source rather than background. The downwind impactor samples show significantly more spread in S/Cl. Particles sized $<1.8 \mu\text{m}$ fall within the 'near-vent bulk aerosol' area, while particles 1.8-10 μm have a much lower S/Cl and fall closer to the seawater mixing line. The 1.8-10 μm size bin is therefore interpreted to contain a much higher proportion of non-volcanic aerosol, although some plume contribution is not excluded. Similarly, the source of PM sized $>10 \mu\text{m}$ appears to be highly mixed.

Figure SM2. Size-resolved chemical composition of aerosol (major and trace species) collected near the eruptive vent ('near-vent', ~5km from emission source) and in a more dilute plume in Reykjavík (~100km from emission source). Note that S and Cl concentrations in near-vent samples 0.1, 0.18 and >18 μm are much higher than in other size bins. They have been divided by 5 to improve the visual clarity of the plot.



References:

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M. Burton, E. Ilyinskaya, M. Hartley, A. La Spina, G. Salerno, E. Bali, S. Barsotti, B. Bergsson, M. A. Pfeffer, H. Kaasalainen, A. Stefansson and T. Thordarson. Mantle source controls gas emissions and impact of Icelandic basaltic eruptions. In review.

Stefansson, A., Stefansson, G., Keller, N.S., Barsotti, S., Sigurdsson, Á., Thorlákssdóttir, S.B., Pfeffer, M.A., Eiríksdóttir, E.S., Jónasdóttir, E.B., von Löwis, S., Gíslason, S.R., 2017. Major impact of volcanic gases on the chemical composition of precipitation in Iceland during the 2014–2015 Holuhraun eruption. *J. Geophys. Res. Atmospheres* 122, 2015JD024093. doi:10.1002/2015JD024093