**Table S1**

Excel file ‘Table\_S1.xlsx. Information about instrumentation, data completeness, data exclusion, etc, for each SO2 and PM monitoring station. Summary statistics for SO2 (hourly-means), PM10, PM2.5 and PM1 (daily-means) data during the background and eruption periods. SO2 concentration data (µg/m3) reported to 2 s.f. Full raw dataset openly available for download from Environment Agency of Iceland <https://loftgaedi.is/en>.

**Figure S1**

Animated simulation of the volcanic SO2 concentration at ground level for the period 28 – 30 May 2021. The colour scale represents the simulated concentrations at ground level (in µg/m3) but should be treated as only as a qualitative indication of plume presence at ground-level. The simulation was made by the CALPUFF dispersion model that was used operationally during the 2021 Fagradalsfjall eruption by the Icelandic Met Office. A detailed methodology of the dispersion simulations is in Pfeffer et al., (2024). The data presented in Figure S1 are unpublished data by the Icelandic Meteorological Office.

**Figure S2**

Animated simulation of the volcanic SO2 concentration at ground level for the period 18 – 20 July 2021. The colour scale represents the simulated concentrations at ground level (in µg/m3) but should be treated as only as a qualitative indication of plume presence at ground-level. The simulation was made by the CALPUFF dispersion model that was used operationally during the 2021 Fagradalsfjall eruption by the Icelandic Met Office. A detailed methodology of the dispersion simulations is in (Pfeffer et al., 2024). The data presented in Figure S2 are unpublished data by the Icelandic Meteorological Office.

**Figure S3**

Map of the total probability (%) of ground-level SO2 concentrations exceeding the 350 µg/m3 air quality threshold during the 2021 Fagradalsfjall eruption. The map is based on dispersion simulations by the CALPUFF model that was used operationally by the Icelandic Meteorological Office. A detailed methodology of the dispersion simulations is in Pfeffer et al., (2024). The model results are used here for qualitative information about the plume direction (as a yes/no indication of the potential plume presence at ground level) because the model had a reasonable skill in predicting the broad plume direction but a relatively low accuracy in simulating the concentrations of SO2 at ground level (Pfeffer et al., 2024). The data presented in Figure S3 are unpublished data by the Icelandic Meteorological Office.