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Temporal Evolution and Atmospheric Impacts of Tropospheric Volcanic Emissions from In-Situ Measurements and Modelling

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Assessment of the impact of tropospheric volcanic gas and aerosol emissions requires integration of observation and modelling. Knowledge and understanding is rapidly advancing in both areas, particularly due to the development of kinetic plume models of reactive halogen chemistry, and due to recent advances in measurement techniques for collecting in situ measurements of plume physico-chemical properties (i.e. using meteorological balloon and aircraft platforms), as well as a proliferation of remote sensing DOAS measurements. Here, we demonstrate this synergic relationship through model-observation plume studies.

Volcanoes are a large natural source of SO_2 and sulphate to the atmosphere, as is well demonstrated from both observational and model studies. In a recent study that deployed quasi-Lagrangian balloons in emissions at Kilauea volcano, Hawaii, both $H_2O(g)$ and $SO_2(g)$ were measured in situ, in the downwind plume. The observations showed periods of both correlation and anti-correlation between SO_2 and water-vapour, implying the occurrence of both source and sink processes. Co-emission of volcanic H_2O with SO_2 accounts for the correlation. We use a thermodynamic model along the plume transect to assess how H_2O-sulphate interactions might account for H_2O anti-correlation with SO_2 within the plume to elucidate in-plume sulphate formation, both near-vent (as predicted by high-T thermodynamic models) and downwind (as predicted by kinetic models).

Volcanoes are a source of halogens (HBr, HCl) to the atmosphere, and volcanic plumes are highly reactive zones, not only in the high-temperature region near the vent, but also in the downwind plume where autocatalytic chemistry cycles produce reactive halogens such as BrO, first discovered from DOAS observations.
The rapid formation of BrO can be reproduced through modelling which predicts high concentrations (reaching ppbv) on short formation timescales (minutes). Simulations using the PlumeChem model (developed to analyse volcanic plume chemistry during atmospheric dispersion) indicate that Br-content, sunlight and plume-air mixing exert important controls on plume chemistry, in agreement with DOAS observations. NO_x is predicted to play a significant role, accelerating reactive bromine chemistry, and thereby converting NO_x into nitric acid. Thus the simulations support a mechanism for elevated HNO_3 observed in volcanic plumes, and highlight additional impacts of plume chemistry. Furthermore, significant ozone depletion was predicted in the downwind plume that could previously only be compared to limited observations. Ozone depletion has recently been characterised by in-situ aircraft measurements of Redoubt (and Erebus) plumes. Our direct comparison of simulated and observed ozone depletion, in good agreement, supports and further constrains estimates of downwind impacts.

The recent advances in in-situ measurements in downwind plumes enable us to test and further develop process-based models of volcanic plume chemistry and impacts, ultimately on a global scale. Conversely the model studies provide interpretation of observations, predict new phenomena and provide spatial simulations of plume chemistry, essential to guide future measurement-strategies.