



Tropospheric impacts of volcanic halogen emissions: first simulations of reactive halogen chemistry in the Eyjafjallajökull eruption plume

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Volcanic plumes are regions of high chemical reactivity. Instrumented research aircraft that probed the 2010 Icelandic Eyjafjallajökull eruption plume identified in-plume ozone depletion and reactive halogens (Cl, BrO), the latter also detected by satellite. These measurements add to growing evidence that volcanic plumes support rapid reactive halogen chemistry, with predicted impacts including depletion of atmospheric oxidants and mercury deposition. However, attempts to simulate volcanic plume halogen chemistry and predict impacts are subject to considerable uncertainties. e.g. in rate constants for HOBr reactive uptake (see this session: EGU2013-6076), or in the high-temperature initialisation. Model studies attempting to replicate volcanic plume halogen chemistry are restricted by a paucity of field data that is required both for model tuning and verification, hence reported model 'solutions' are not necessarily unique. To this end, the aircraft, ground-based and satellite studies of the Eyjafjallajökull eruption provide a valuable combination of datasets for improving our understanding of plume chemistry and impacts.

Here, PlumeChem simulations of Eyjafjallajökull plume reactive halogen chemistry and impacts are presented and verified by observations for the first time. Observed ozone loss, a function of plume strength and age, is quantitatively reproduced by the model. Magnitudinal agreement to reported downwind BrO and Cl is also shown. The model predicts multi-day impacts, with reactive bromine mainly as BrO, HOBr and BrONO₂ during daytime, and Br₂ and BrCl at night. BrO/SO₂ is reduced in more dispersed plumes due to enhanced partitioning to HOBr, of potential interest to satellite studies of BrO downwind of volcanoes.

Additional predicted impacts of Eyjafjallajökull volcanic plume halogen chemistry include BrO-mediated depletion of HO_x that reduces the rate of SO₂ oxidation to H₂SO₄, hence the formation of sulphate aerosol. The model predicts NO_x is rapidly converted into nitric acid (via BrONO₂). Such HNO₃-formation might contribute towards new particle formation, noting reported very high in-plume particle nucleation rates in Eyjafjallajökull plume. Thus, plume halogen chemistry influences on aerosol formation and growth are emphasized regarding studies of climatic and health impacts of volcanic aerosol.

As the plume disperses, in-plume ozone concentrations partially recover due to entrainment of O₃-rich background air. However, the cumulative net impact on ozone depletion continues. Whilst the global tropospheric impact of Eyjafjallajökull is small, up-scaling of the model findings in the context of present day global volcanic degassing and recent historic eruptions indicates potential for significant impacts of global volcanic halogen emissions on tropospheric ozone, particularly during periods of enhanced volcanic activity.

Notably, this model-observation study of Eyjafjallajökull plume exhibits contrasts to a related model-observation study that quantified ozone loss in Redoubt volcano eruption plume (Kelly et al., JVGR in press). Meteorological and volcanological causes for these differences in plume halogen evolution (hence impacts) are discussed. This has implications for wider atmospheric modelling efforts to quantify global impacts from volcanic halogen emissions and highlights the useful role of fully-flexible and computationally inexpensive models such as PlumeChem to inform larger (regional or global) model studies regarding model initialisation and particularly near-source plume chemistry.