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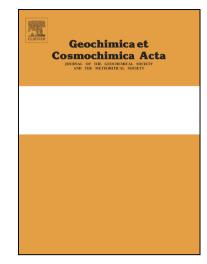
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ACCEPTED MANUSCRIPT

The primary volcanic aerosol emission from Mt Etna: size-resolved particles with SO₂ and role in plume reactive halogen chemistry

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Abstract

Volcanoes are an important source of aerosols to the troposphere. Within minutes after emission, volcanic plume aerosol catalyses conversion of co-emitted HBr, HCl into highly reactive halogens (e.g. BrO, OCIO) through chemical cycles that cause substantial ozone depletion in the dispersing downwind plume.

This study quantifies the sub-to-supramicron primary volcanic aerosol emission (0.2-5 μ m diameter) and its role in this process. An in-situ ground-based study at Mt Etna (Italy) during passive degassing co-deployed an optical particle counter and Multi-Gas SO_2 sensors at high time resolution (0.1 Hz) enabling to characterize the aerosol number, size-distribution and emission flux.

A tri-modal volcanic aerosol size distribution was found, to which lognormal distributions are fitted. Total particle volume correlates to SO_2 (as a plume tracer). The measured particle volume: SO_2 ratio equates to a sulfate: SO_2 ratio of 1-2 % at the observed meteorological conditions (40% Relative Humidity). A particle mass flux of 0.7 kg s⁻¹ is calculated for the measured Mt Etna SO_2 flux of 1950 tonnes/day.

A numerical plume atmospheric chemistry model is used to simulate the role of the hygroscopic primary aerosol surface area and its humidity dependence on volcanic plume BrO and OCIO chemistry. As well as predicting volcanic BrO formation and O_3 depletion, the model achieves $OCIO/SO_2$ in broad quantitative agreement with recently reported Mt Etna observations, with a predicted maximum a few minutes downwind. In addition to humidity – that enhances aerosols surface area for halogen cycling – background ozone is predicted to be an important control on

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