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Halogen activation in the plume of Masaya volcano

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Volcanoes are a main source for several reactive atmospheric trace gases including sulfur and halogen containing species. The detailed understanding of volcanic plume chemistry is needed to extract information from gas measurements on subsurface processes. Therefore, this knowledge is essential when using gas data as a monitoring tool for volcanic activity. The reactive bromine monoxide (BrO) is of particular interest because BrO, as well as SO_2, is readily measurable from safe distance by spectroscopic remote sensing techniques. Already today, BrO/SO_2 time series of 10 years and more exist for several volcanoes. However, bromine is assumed to be emitted as HBr and BrO is thought to be formed in the plume by a multiphase reaction mechanism with Br_2as an intermediate product. The abundance of BrO changes as a function of the distance from the vent as well as of the spatial position in the plume.

In this study, we present observations of the oxidation of bromine, chlorine and iodine during the first 11 minutes after emission in the plume of Santiago Crater of Masaya volcano (Nicaragua) obtained by ground- and UAV-based measurements, the later ones were particularly essential for otherwise inaccessible plume ages. In situ sampling by alkaline traps and gas diffusion denuders enabled observations of the reactive halogen (e.g., Br_2, BrCl, BrO, HOBr) to total halogen and sulfur ratios at various distances from the vent. Additional reactive halogen to sulfur ratios were furthermore obtained by simultaneous application of gas diffusion denuder sampling and electrochemical SO_2 sensors. The in situ data is complemented by spectroscopic UV-measurements taken by 1.4 km downwind permanent installed instruments of the NOVAC network and analyzed with the DOAS technique. The reactive bromine to total bromine molar ratio increased from 0.20 $\pm$ 0.13 at the crater rim to 0.76 $\pm$ 0.26 at further downwind (2.8 km). Chlorine only showed an increase of the reactive molar fraction from $(2.7\pm0.7)\times10^{-4}$ to $(11\pm3)\times10^{-4}$ in the first 750 m while the reactive fraction of iodine increases from about 0.3 to about 0.9 on the first 2.8 km. BrO/SO_2 molar ratios increased from 0.95×10^{-5} at an estimated plume age of 1.4 minutes to 3.7×10^{-5} at 11.1 minutes and BrO contributes to about 10% of the reactive bromine species during that plume age.

The obtained field data was compared with simulations of an atmospheric box model (CAABA/MECCA) that was initiated with the output of a thermodynamic equilibrium chemistry model (HSC). Different model parameters were varied in order to investigate scenarios, which enable the reproduction of the field observations. The model runs, which best produced our observations, will be shown and the most important model parameter will be discussed.