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Volcanic plumes: BrO formation during the first 100 minutes after emission

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The main volatile species emitted by volcanoes are H_2O, CO_2 and SO_2 in order of magnitude. Significant amounts of HCl are also emitted along with a minor fraction of other hydrogen halides (HX, $X = F, Cl, Br, I$). Halogen radicals can be formed in the plume from HX. E.g., BrO is produced in the volcanic plume from the emitted HBr in an auto-catalytic reaction cycle similar to the "Bromine-Explosion" known from the Arctic and Antarctic tropospheric chemistry. Reactive bromine species (and probably analogous chlorine species) are known to have a strong influence on the oxidation capacity of the atmosphere. The chemical abundance of BrO in volcanic plumes is commonly described by the BrO/SO_2 ratio, as SO_2 is chemically inert on time scales of hours, and can therefore be used as a tracer for dilution of volcanic gas. Based on measured BrO/SO_2 ratios, improved numerical models of the chemical plume evolution could be used to estimate the total amount of bromine emitted by the volcano as well as the impact of volcanic emissions on the atmosphere.
BrO/SO_2 ratios will be presented which were measured at Mt. Etna during July 2008 and July 2009 with differential optical absorption spectroscopy (DOAS). These measurements were performed simultaneously with 1-3 stationary Multi-Axis DOAS instruments (MAX-DOAS) and car based traverses. They constitute the first capture of the evolution of BrO/SO_2 ratios in a volcanic plume in the first two hours after emission, and are the hitherto most comprehensive data set of BrO/SO_2 ratios from any single volcano. Different evolutions of ratios for both measurement campaigns are observed. During the July 2008 measurements, a small but consistent increase in BrO/SO_2 with increasing plume age was observed in the first hour, with a maximum value of $BrO/SO_2 = 1.2 \cdot 10^{-4}$ found after 60 minutes. In contrast, measurements during July 2009 generally show smaller BrO/SO_2 ratios. After an initial increase from 0 to $0.65 \cdot 10^{-4}$ during the first 5 minutes, a decrease of the ratio appears to occur. Previously published chemical model runs do not adequately represent the encountered boundary conditions and measurements, and overestimate the observed evolution of BrO/SO_2 ratio at plume ages greater than 5 minutes by a factor of 3. Thus additional chemical model runs are presented which are based on the new data set. The impact of the new insights is discussed.