Geophysical Research Abstracts
Vol. 14, EGU2012-10184-2, 2012
EGU General Assembly 2012

Sulfur isotopic composition of fumaroles and plume at Etna volcano (Italy)

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Mount Etna is one of the most active volcanoes worldwide and is among the main dischargers of SO_2 in atmosphere. The isotopic signature of the outgassed sulfur carries information on mantle source of magmas, crustal and hydrothermal contributions to volcanic fluids, and physico-chemical conditions inside the plumbing system. With the aim to asses the not-well constrained sulfur isotopic composition at Etna, this research proposes new measurements of δ^{34}S ratio in both plume and fumarolic gases. The peculiar geochemical features of plume and high temperature gases (strongly air-diluted) have required the development of proper sampling methods: high temperature gases were sampled in a pre-evacuated 4l glass bottle where we injected 100 ml of 1M NaOH solution; plume gases were collected through a bubbler properly constructed to allow the gas to dissolve in an alkaline solution (1M NaOH). Collected samples display δ^{34}S values between -1 and +4‰ vs. V-CDT. Within this range, low temperature fumaroles show δ^{34}S-values systematically higher than high temperature gases and plume. We deduce that such values do not reflect the pristine isotopic composition of S released by magma, but are reasonably affected by post-magmatic processes. On the contrary, we infer that the average δ^{34}S measured in high temperature fumarole and in plume gases is representative of the magmatic signature.

Our data of sulfur isotopic composition are compared to those available in literature on S dissolved in primitive melt inclusions hosted in olivines from 2002 lava and in whole rocks erupted in the past 2000 years, showing that δ^{34}S of magmatic gases is lower than that in the melt. Modeling requires assessment of the fractionation factor between S dissolved in the melt and exsolved in the gas phase ($\alpha_{gas-melt}$). It depends on the speciation of sulfur in both the melt and gas phases, which is in turn strictly related to the oxygen fugacity ($f\mathrm{O}_2$) of the magma and to temperature and water fugacity (f_{H2O}) at which the degassing occurs. Within the range of redox conditions reported in literature for Etnean basalts, isotopic fractionation could lead to either ^{34}S depletion or enrichment of the gaseous phase. Nevertheless, the most recent measurements on the oxidation state of sulfur in basaltic inclusions hosted in olivines indicate that nearly all S is dissolved as sulfate (S^{6+}), that can be possible only in oxidized magmatic systems. Under these conditions the exsolved gaseous phase is depleted with respect to the melt, in accordance with our data. In this case, the proposed model of isotopic fractionation due to magma degassing (Rayleigh-type) is able to fit both gas and melt data and to constrain the Etnean magmatic δ^{34}S to +1±1.5‰ vs. V-CDT. Related inferences on the plumbing system are also discussed.