Origin of natural hydrogen discharging from geothermal fumarolic emissions in Monterotondo, Tuscany region, Italy

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Northern Appenines including Tuscany region is a complex tectonic environment where the contractional deformations of the Cretaceous-Miocene collisional tectonism were overprinted by the Late Miocene-Quaternary post-collisional magmatism and back-arc extension. Emplacement of shallow-level magmatic intrusions produced a wide thermal anomaly into the Tuscan crust and triggered the development of diffuse thermal aureoles and associated hydrothermal systems. Two main geothermal systems exist: the "shallow" hosted in the Mesozoic evaporite-carbonate units (about 0.7 – 1.0 km with temperature from 150°C to 260°C) and the "deep" localized in the Paleozoic metamorphic succession and Neogene granitoids (about 2.5 – 4.0 km with temperatures exceeding 300°C). Circulating geothermal fluids likely facilitate water-rock interaction processes which produce several types of fluids discharging at the surface.

The sampled fumarolic gases are chracterized by locally high H_2 concenteration (0.2-10.3 vol%), elevated H_2/CH_4 (up to 4.8) with wide ranges of CO_2 (15-78 vol%) and N_2 (17.5-80 vol%). $\delta^{13}C$ values of CO_2 and CH_4 range from -0.9 to -4.2% and from -22.1 and -29.6%, respectively. Moreover, δD values of hydrogen and methane range from -415. to -558.% and from -171.75 to -185.%, respectively.

The CO2-CH4 and CH4-H2 isotopic systematics suggest a volcanic-magmatic origin of the fumarolic CO2. The studied fumarolic gases have the isotopic signatures comparable to those originated in oceanic settings; they retain the isotopic signature of hydrogen produced by high-temperature (~ 350-450 °C) water-rock interactions. The highest H2 content are located above the granitic intrusion that forms the larderello geothermal province. Potential sources of H2 are so numerous: oxydation of the granite itself, the ophiolitic nappes heated by the granite intrusion or the radiolyse also due to the granite body. Or a mix of these processes since H2 is likely produced by interaction between overpressured fluids circulating in the deep geothermal system (Tuscan metamorphic complex) which migrated through the deep-seated faults and triggered high-temperature serpentinization of the ophiolitic basement hosted in the Tuscan ligurian units.

Mots-Clés: Tuscany, Fumarolic gas, Hydrogen, Isotope geochemistry, Serpentinization, Ligurian ophiolites.

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