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Preliminary geochemical characterization of gas manifestations in North Macedonia

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Like most of the Balkan Peninsula, North Macedonia is a geodynamically active area. As such it has many hydrothermal features and gas manifestations. Until now, no systematic study about the geochemical characterization of the geogenic gases was made before in this country. In August 2019, 24 gas samples were collected in the study area. All, except one collected at Duvalo (soil gas), are gases bubbling or dissolved in thermomineral waters (temperatures from 12 to 66 °C). They were analysed in the laboratory for their chemical (He, Ne, Ar, O₂, N₂, H₂, H₂S, CH₄ and CO₂) and isotopic composition ($\delta^{13}\text{C-CO}_2$, $\delta^{13}\text{C-CH}_4$, $\delta^2\text{H-CH}_4$ and R/R_A). Most of the gases have CO₂ as the main component (400-998,000 ppm) while the remaining are enriched in N₂ (1300-950,000 ppm). Helium ranges from 0.3 to 2560 ppm while CH₄ from 1.6 to 20,200 ppm. R/R_A and $^4\text{He}/^{20}\text{Ne}$ ratios indicate a generally low atmospheric contamination, a prevailing crustal contribution and mantle contributions between 1 and 20% considering a MORB endmember. The highest mantle contributions are found in the SE part of the country very close to the sites that show the highest R/R_A values in continental Greece [1]. This area is characterised by extensional tectonics and Plio-Pleistocene volcanism. A quite high mantle contribution (about 15%) is also found in two manifestations in the NW part of the country along a main normal fault system. With the exception of the sample of Smokvica, which has very low CO₂ (1400 ppm) and $\delta^{13}\text{C-CO}_2$ (-15.7 ‰ V-PDB), all free gases show a relatively narrow range in $\delta^{13}\text{C-CO}_2$ values (-4.6 to +1.0 ‰ V-PDB) indicating the mixing between a mantle and a carbonate rock source. The isotope composition allows us to assign the CH₄ origin to three sources. The largest group can be attributed to a hydrothermal origin ($\delta^{13}\text{C-CH}_4$ around -20 ‰ V-PDB and $\delta^2\text{H-CH}_4$ around -100‰). Three samples collected in the SW part of the country have a thermogenic origin ($\delta^{13}\text{C-CH}_4$ around -35 ‰ V-PDB and $\delta^2\text{H-CH}_4$ around -160‰ V-SMOW). Finally, one sample (Smokvica) with the highest values ($\delta^{13}\text{C-CH}_4$ -7.2 ‰ V-PDB and $\delta^2\text{H-CH}_4$ -80‰ V-SMOW) may be attributed to abiogenic processes in a continental serpentinization environment or to methane oxidation.

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