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Long-lived Radioactive Elements and REE as Fingerprints of Deep Groundwater Flow

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Groundwater originating from great depths provide a valuable geochemical sampling medium for exploring the development of the Earth's crust, geological, and hydrogeological resources. This particularly applies to sites of natural springs, where favorable hydrogeological conditions enabled regional discharge. Despite the numerous occurrences of mineral and thermal waters in Serbia, the current understanding of the regional groundwater flow is associated with many open questions that need to be addressed. From a geological standpoint, Serbia is part of the Alpine-Mediterranean mountain belt. From the middle of the Mesozoic to the present, this area underwent processes of subduction, collision, and extensions with accompanying voluminous magmatism and volcanism. As a result of the mentioned geodynamic events, the Serbian territory was a zone of intensive tectonomagmatic processes which had a significant impact on the formation of the hydrogeological structures for forming groundwater enriched with specific elements and elevated temperatures.

Understanding groundwater origin and characterization of a deep circulation is a big challenge since the groundwater pathways and aqueous chemistry are significantly influenced by various factors. To contribute to the characterization of the hydrogeological systems in which the mineral and thermal waters of Serbia are formed, a general hydrochemical study was conducted. During this research 190 of the most significant sources of mineral and thermal waters were sampled, belonging to different geological (geotectonic) units all over Serbia. The applied hydrochemical approach of recognition of deep circulation patterns is based on an analysis of rare earth elements (REE) and natural radioactivity. REE and long-lived radionuclides ^{40}K, ^{238}U, ^{232}Th, ^{226,228}Ra, gross alpha, and beta radioactivity, have proven to be significant fingerprints of water-rock interaction as well as groundwater flow tracers.

The integrated approach of the hydrogeochemical analysis and multivariate statistical method, including spatial mapping of obtained results, was an important process for meaningful interpretation of the data set. The applied approach summarized the complex hydrochemical properties on a general level defining specific hydrochemical fingerprints of hydrogeological systems with distinct geochemical characteristics and flow patterns. Geochemical behavior of natural tracers (REE) and radioactivity contributed to further characterization of deep hydrogeological systems in basins structures, hard rocks (igneous and metamorphic rocks), as well

as carbonate environments.

Rare-earth element data (including abundances and fractionation patterns along with anomalies of Ce and Eu and interelement ratios), relationships of U and Th as elements with different geochemical behavior, and the content of Ra in groundwaters have been singled out as important indicators of deep hydrogeological systems. The results showed that the isolated regional hydrogeological systems are in the function of significant tectonic structures/dislocations, but also hydrogeological characteristics and circulation conditions. Further use of the proposed methodology will provide important data from the assessment of the origin of hydro-geofluids in Serbia and contribute to the wider picture in the understanding of the hydrogeological evolution of regional groundwater flow.

Keywords: natural radioactivity, rare earth elements, hydrogeochemical fingerprints, regional groundwater flow