

2.O b.y old natural nuclear wastes: the natural nuclear reactors in Gabon

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Two billions years ago, the increase of oxygen in atmosphere and the high $^{235}\text{U}/^{238}\text{U}$ uranium ratio(>3%) allowed the formation of natural nuclear reactors on the earth. The particular geochemical conditions (low REE and B contents which are “poison” for neutrons) and the accumulation of high-grade uranium ores at Oklo initiated fission reactions which sustained for 100 000 to 500 000 years. These reactors are now considered to be good natural analogues for nuclear waste disposals. Their preservation during such a long period of time is mainly due to the geological stability of the site, the occurrence of clays surrounding the reactors and acting as an impermeable shield and the occurrence of organic matters that maintained the environment in reducing conditions, favourable for the stability of uraninite. Hydrogeochemical studies and modelling have shown the complexity of the geochemical system at Oklo (deep reactors: 100 - 400m) and Bangombé (reactor located close to the surface: 12m). It has been shown that various processes, including adsorption and precipitation, are involved in the retention of fission products and actinides. The efficiency of these processes depends on the mineralogy of the host rocks (occurrence of clays, oxides, phosphates...) and the chemistry of the groundwater. It can be shown that these parameters change with time and from one area to another. Consequently, it is of great concern to better understand the mechanisms involved during the water-rock interactions that control the water chemistry and consequently the behaviour of fission products and actinides in the natural environment for a long period of time. In this respect, the natural nuclear reactors in Gabon are ideal to illustrate the effects of time on the evolution of the geochemical processes involved in the retention/migration of fission products and actinides. In the reactors and their vicinity, the occurrence of many chemical elements with abnormal isotopic compositions (fission products and elements with high section de capture) give the unique opportunity to trace in detail the behaviour of fission products and actinides during the various geochemical processes.

Hydrological stability in carbonate aquifers over the last 250kyr as reflected by $^{234}\text{U}/^{238}\text{U}$ in groundwater, speleothems and tufa

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The main regional aquifers in the Eastern Mediterranean are set in carbonate rocks dating from Jurassic to Upper Cretaceous. $^{234}\text{U}/^{238}\text{U}$ activity ratios in groundwater emerging from these aquifers vary over a wide range, from 1.0-2.5. However, the range of values obtained in the water in each of the three regions sampled (Mt. Hermon, Galilee, and the Judea Mountains) exhibits a narrow range of values (2.21-2.48, 0.99-1.07 and 1.01-1.13 respectively). Thus, these water sources can be characterized by their $^{234}\text{U}/^{238}\text{U}$ activity ratios. Present-day speleothems and tufa associated with these waters exhibit activity ratios similar to the water from which they precipitated.

Paleo tufa and speleothems within each region were dated using the U- ^{230}Th (TIMS) method and found to cover the age range of the last 250kyr. The $^{234}\text{U}/^{238}\text{U}$ activity ratios obtained from these older samples remain similar to recent values but have a wider range. Whereas the wider range reflects the overall variation in the $^{234}\text{U}/^{238}\text{U}$ activity ratios over several glacial-interglacial intervals (Ayalon et al., 1999), the differences between the regions, remains similar to that found today. This suggests that the $^{234}\text{U}/^{238}\text{U}$ activity ratio in carbonate aquifers is determined mainly by the local hydrological and aquifer conditions (rock and vegetation type, water paths, residence time) and less by the changes in temperature and precipitation amount.

The $^{234}\text{U}/^{238}\text{U}$ ratios in the water and secondary carbonates were measured by MC-ICP-MS and compared with values derived by TIMS. Excellent precision was obtained by the MC-ICP-MS (NU instrument) with values similar within error to those determined by TIMS.

Reference

Ayalon A., Bar-Matthews M. and Kaufman A. (1999).
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