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Uranium and strontium isotopes in recent and interglacial tufa in Krka National Park (Croatia)

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The potential of some non-traditional isotopes (U, Sr) to elucidate the environmental influences on tufa formation and to estimate the proportion of authigenic and detrital carbonate in tufa precipitated from the Krka River (Croatia) was explored. The spatial variability of geochemical (Mg/Ca, Sr/Ca, U/Ca) and isotopic ($\delta^{18}O$, $\delta^{13}C$, $^{87/86}Sr$, $\delta^{88}Sr$, $\delta^{238}U$, and $^{234}U/^{238}U$ activity ratios) parameters was analysed in recent and interglacial tufa at the main cascades and compared to those in the contributing sources (water, bedrock, soil).

A general comparison with recent tufa showed that old tufa had on average higher $\delta^{13}C$, $\delta^{18}O$ and $\delta^{88}Sr$ values of carbonate, $\delta^{13}C$ values of sedimentary organic carbon (C_{org}), carbonate-bound Mg concentration and $\delta^{238}U$ values of non-carbonate fraction. On average lower values in old tufa were measured for the concentration of Sr and C_{org}, the C_{org}:N ratio, non-carbonate mineral fraction and U concentrations (bulk and carbonate-bound fraction). The $^{234}U/^{238}U$ activity ratios, $\delta^{238}U$ values of carbonate and $^{87/86}Sr$ ratios were similar.

The concentration of C_{org} in old samples decreased during diagenetic decomposition, when the ^{13}C-depleted C was preferentially lost, explaining the increased $\delta^{13}C$ of C_{org}. Diagenetic processes can also explain the decreased Corg:N ratio in old tufa, where the mineralisation of organic matter inside the tufa barrage resulted in increased concentrations of ammonia in interstitial water, which remained adsorbed on mineral (in particular clay or Fe hydroxide) surfaces. The lower concentration of U in old tufa can be reasonably explained by increased U concentrations in recent Krka River from the U-enriched industrial discharge in the upper reaches of the river and lower amount of non-carbonate mineral fraction, which would indicate less terrestrial input through surface runoff at the locus of tufa precipitation in the last interglacial period. The downward decrease of the non-carbonate $\delta^{238}U$ values in the interglacial tufa profile is explained by the increasing input of strongly ^{238}U depleted detrital material originating from weathering of Cretaceous limestone outcropping in the catchment area, related to the hydrological changes since the last interglacial period, which are reflected also in $\delta^{88}Sr$ values of tufa.