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$^{234}U/^{238}U$ Disequilibrium along stylolitic discontinuities in deep Mesozoic limestone formations of the Eastern Paris basin: evidence for discrete uranium mobility over the last 1–2 million years

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Abstract. The ($^{234}U/^{238}$) equilibrium state of borehole core samples from the deep, low-permeability limestone formations surrounding the target argilite layer of the Meuse/Haute-Marne experimental site of the French agency for nuclear waste management -ANDRA- (Agence nationale pour la gestion des déchets radioactifs) was examined to improve understanding of naturally occurring radionuclide behaviour in such geological settings. Highly precise, accurate MC-ICP-MS measurements of the ($^{234}U/^{238}U$) activity ratio show that limestone samples characterised by pressure dissolution structures (stylolites or dissolution seams) display systematic ($^{234}U/^{238}U$) disequilibria, while the pristine carbonate samples remain in the secular equilibrium state. The systematic feature is observed throughout the zones marked by pressure dissolution structures: (i) the material within the seams shows a deficit of ^{234}U over ^{238}U (($^{234}U/^{238}U$) down to 0.80) and (ii) the surrounding carbonate matrix is characterised by an activity ratio greater than unity (up to 1.05). These results highlight a centimetric-scale uranium remobilisation in the limestone formations along these sub-horizontal seams. Although their nature and modalities are not fully understood, the driving processes responsible for these disequilibria were active during the last 1–2 Ma.

Keywords: uranium isotopes, multiple-collector ICP-MS, waste management, remobilisation, migration

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