U-series disequilibria in deep fracture zone in the Vienne granitoids as an indicator of uranium migration processes

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Uranium concentration and activity ratios ($^{234}$U/$^{238}$U and $^{230}$Th/$^{234}$U) in groundwater, fracture coatings and adjoining rock matrix were studied in the Vienne granitoids, France. The aim was to develop and test a conceptual model of the groundwater flow system. The small to moderate disequilibrium ($1.87 < ^{234}$U/$^{238}$U < 3.36) observed in the deep groundwater, together with very low uranium concentration, is characteristic of reducing conditions. It is very likely that deep groundwaters with uranium contents as low as 0.022 ppb, underwent activity ratio increase due to the alpha-recoil process, while shallow groundwaters, with uranium content close to 24 ppb, result from preferential $^{234}$U-solution processes. Activity ratios as a function of distance from fractures surface (sample at 342m) show clear absolute uranium release up to 5 to 6 cm's from the fracture surface. Both $^{230}$Th/$^{234}$U and $^{234}$U/$^{238}$U profile indicates episodic $^{234}$U mass flow events within the last 350,000 years. A working hypothese to relate observed absolute uranium release and isotopic fractionation between isotopes $^{234}$U and $^{238}$U is based on the radioactive decay induced oxidation of $^{234}$U to U(VI), that is the more soluble oxidation state of uranium. In this situation the oxygen concentration in groundwater plays an important role. If there is much oxygen it can actually oxidise and release the bulk of uranium which would not lead to marked isotopic fractionation. If there are
small amounts of oxygen, the already oxidised $^{234}$U is easier to remove leading to clear isotopic fractionation. If finally there is very little or no oxygen, it would have no effect even for $^{234}$U in which case there would not be absolute release nor preferential $^{234}$U release.