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Assessing the role of barite to control ^{226}Ra long term behavior in McClean tailings - Saskatchewan, Canada

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Radium-226 is the predominant radionuclide of most NORM (Naturally Occurring Radioactive Material) waste, and is of special interest for the Canadian U-mines with high grade ore. The ore treated at McClean lake mill - Saskatchewan, Canada – has activities averaging 2200 Bq/g ^{238}U and ^{226}Ra, and the recovery rate for U is 99%. U mining mill tailings are sulfate-rich low-level radioactive waste containing ^{226}Ra, one of the main contaminants of interest in tailings from U-mines, with activities up to 3000 Bq/g. In order to develop a robust modelling of the long-term behavior of ^{226}Ra, a good comprehension of the geochemical retention mechanisms at work and a thorough knowledge of the composition and dynamics of the tailings are required. At the McClean mill $BaCl_2$ is added to precipitate ^{226}Ra in barite under the form of a solid-solution. In the tailings, ^{226}Ra is generally considered as sequestered in barite and sorbed on ferrihydrite. To assess the stability of ^{226}Ra in the tailings after $BaCl_2$ addition, an extensive study of the potential ^{226}Ra-bearing phases in tailings samples was performed. The work is conducted to ensure that the tailings have no significant effects on the downstream environment which remains protected over the long term. The combined results of chemistry, mineralogy, SEM/EDS and alpha-autoradiography analyses supplemented with thermodynamic modelling confirm that barite is the main ^{226}Ra binding-phase, in agreement with the ^{226}Ra concentration measured in pore water (6 Bq/L). The equilibrium with a $(Ba,Ra)SO_4$ solid-solution controls the ^{226}Ra concentration now and over the long term through a distribution coefficient with an initial value reflecting a coprecipitation mechanism (D = 1.5-2.2), and that evolves towards one of pure recrystallization (D = 0.1-1).