Behavior of uranium isotopes in shallow aquifers of southern Québec, Canada

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A multi-isotopic study was initiated in order to quantify the groundwater resources available in a shallow aquifer of southern Québec. Uranium content and its isotopes ^{234}U and ^{238}U, which are partially dependent of redox conditions and physical/lithological characteristics of the aquifers and their recharge were measured. In specific cases, $^{234}U/^{238}U$ activity ratios can give information on the residence time of groundwater, a fundamental piece of information for a correct estimation of groundwater sustainability.

The target area is the lower portion of the Bécancour River watershed located between Montréal and Québec. Groundwater flows in shallow sands intercalated with clays of the Champlain Sea (Holocene age) and deeper fractured carbonates of Ordovician age of the St. Lawrence Lowlands. Water chemistry is dominantly bicarbonate.

Two transects (15 wells) were preliminary selected. The first follows the main flow path from the Appalachian Mts. (the main recharge area) downstream to the St. Lawrence River. The second transect is perpendicular to the first one and contains more mineralized waters. Along the flow path, there is a clear decreasing trend of the $^{234}U/^{238}U$ activity ratio, from a maximum of 3.12 measured close to the recharge to a value of 1.14, on the plain next to the St. Lawrence River.

During decay of ^{238}U to ^{234}Th, an α particle is emitted. The recoil energy is sufficiently important for the mineral to eject ^{234}Th to groundwater or to weak mineral sites were ^{234}U will be produced, making it easily leachable. Consequently, recharging groundwater would be enriched in ^{234}U when compared to ^{238}U and this mechanism might explain the high $^{234}U/^{238}U$ activity ratios observed close to the recharge areas.

The observed decrease of the $^{234}U/^{238}U$ ratio along the flow path could be interpreted by the radioactive decay of the excess of ^{234}U compared to that of ^{238}U at the ^{234}U time scale (10^6 yrs). However, a recent noble gas survey shows clearly that groundwater in the recharge area contains tritiogenic ^{3}He with ages younger than 20 years. A different process should be evoked to explain the isotopic fractionation of $^{234}U/^{238}U$. There is a clear relation between the alkalinity of waters (and the HCO_3^- content) and the $^{234}U/^{238}U$ activity ratio. This relation suggests that the mobility of uranium might be related to HCO_3^- and CO_3^{2-} ions complex. It is known that carbonates and hydroxyls could be efficient agents of complexation inducing higher mobility of ^{234}U after the “recoil”. Thus the high activity ratios observed could be related to the enhanced mobility of ^{234}U in bicarbonate waters.

Low energy absorption edges of NS3 glass, albite and silicon investigated using x-ray Raman scattering

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X-ray Raman Scattering (XRS)

The investigation of processes of geological relevance is directly connected to environments of extreme conditions, e.g., high pressure and high temperature. For in situ studies of materials under such conditions one needs to use special sample chambers with highly absorbing sample environments such as diamond anvil cells, which prevent the use of electron or soft x-ray techniques like EELS or XANES, combined with laser or resistive heating.

XRS is a non resonant inelastic photon-in-photon-out scattering process. The incoming high energy photons are scattered by core-hole electrons, exciting them to unoccupied states by transferring a relatively small amount of energy [1]. The application of hard x-rays makes x-ray Raman scattering a unique tool to investigate low energy absorption edges for binding energies between 10 eV and 2 keV of low Z elements like silicon, sodium, iron or oxygen under geologically relevant conditions.

XRS is very sensitive to changes of the electronic and local atomic structure, e.g., the oxidation or spin state of the studied atom. It provides the similar information as soft x-ray absorption and electron energy loss spectroscopy.

NS3 glass, albite and silicon

Silicon is the subject of many current studies. These include silicate melts and glasses due to their geological relevance, as well as pure silicon [2,3]. We present first results of an XRS study of the sodium K-edge of dry and water bearing NS3 glass ($Na_2Si_3O_7$) and albite ($NaAlSi_3O_8$) as well as results on the aluminum L-edge of albite. Furthermore we show recent in situ measurements of pure silicon at the Si L-edge at pressures up to 20 GPa.

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