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Migration of radioactive iodine, technetium and selenium in soils in response to gradients in near-surface redox potentials

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The proposed geological disposal of radioactive waste in several countries has led to the consideration of risks arising from the long-term migration of radionuclides from sub-surface repositories to the biosphere. The relative risks of specific radionuclides migrating to the surface environment over long (>1000 year) timescales can be ranked on the basis of their physical half lives and their tendencies to adsorb to geological materials. Long-lived and relatively poorly sorbed radionuclides within radioactive wastes are more likely to 'break through' to the surface environment, though still in very small quantities. Radionuclides with such characteristics include I-129, Tc-99 and Se-79. Consequently, these have been the focus of studies designed to provide information to help improve models of radionuclide migration and impact in the context of radioactive waste disposal.

A critical step in the movement of radionuclides from the sub-surface environment to the biosphere is the transition from saturated to partially saturated conditions within the soil profile. The proximity to the surface of this interface depends on the type of soil and biome being considered. It is also liable to fluctuate according to seasonal, or even daily, variations in rainfall and evapotranspiration. Both temporal and spatial changes in soil wetting can lead to significant fluctuations in oxidation-reduction potential and, hence, the speciation of elements which are redox-sensitive, including iodine, technetium and selenium. This leads to substantial variations in the sorption of these elements in the soil and adds to uncertainties associated with risk calculations.

A series of soil column and 'mini-column' experiments has been carried out to assess the sorption and migration of I-129, Tc-99 and Se-79 (or isotopic surrogates) in re-

sponse to gradients in soil redox potential. In the case of selenium and technetium, permanently saturated subsoils, with low (negative) redox potentials, have been found to present a barrier to vertical migration within soil columns. However, it has been observed consistently that small quantities of Tc-99 and Se-75 are able to migrate past this barrier, into oxic surface soil and eventually into plant roots. There is evidence that processes such as biological transformation and translocation may enhance this small degree of transport which may be significant over the time scales considered in radioactive waste safety studies. Iodine-125 has been observed to migrate readily in a reduced form (iodide) within the anoxic sub-surface, although significant accumulation occurs at the boundary between anoxic and oxic soil as oxidation to the iodate form occurs. This parallels observations of iodine in marine sediments in which accumulation occurs at the oxic-post oxic boundary. For I-129, with a 15.7 million year half life, this represents a potentially important accumulation mechanism which could result in significant sinks of radioactive iodine in soils with appropriate hydrological conditions.

Our laboratory studies have demonstrated that soil redox potential is a critical factor which must be taken into account when evaluating the long term biogeochemical cycling of radionuclides in the biosphere. Future studies are aimed at using naturally occurring isotopic surrogates to investigate accumulation of redox sensitive radionuclides at oxic/anoxic boundaries in field soils.