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Ra-226 in Bone of Mink (*Mustela vison*) and Otter (*Lutra canadensis*) Taken Near U Workings at Elliot Lake, Canada, and from Reference Areas, with Calculation of Transfer Parameters

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The City of Elliot Lake was an important focus of Canadian uranium operations from 1955 until 1996 when the last mine closed. Uranium mines and mills were located in the upper and middle sections of the Serpent River watershed that drains to the North Channel of Lake Huron (Figure 1) at Serpent Harbour (~35 km S). Company records indicate forty years of mining and milling U ore left ~150 x 10^6 tonnes of acid-generating wastes (tailings and rock) covering ~600 ha in the watershed (see Clulow 1995). The wastes contain metals and radionuclides that leach out or are released as particulates or gases and may be taken up by local biota, including man. The watershed has been intensely monitored since the 1960s. Changes in water quality (principally pH and dissolved ^{226}Ra levels) reflect changes in mining practices, levels of mining activity, and in the treatment of effluents during this period. Mean dissolved ^{226}Ra levels in the lower Serpent River, just upstream from its mouth, declined from about 250 mBq L^{-1} (1969 – 1975) to between 50 and 100 mBq L^{-1} in the period from 1980 to the mid 1990s (when monitoring was discontinued).

In our study of environmental radioactivity of the Elliot Lake region, ^{226}Ra levels in local mink and otter bones (sampled in 1986 and 1997) were compared to others taken from areas without U operations. These comparisons provide insight into ^{226}Ra levels in mammalian secondary consumers in an ecosystem containing U mines and mills (compared to control levels) and change related to decline of U operations (by comparison of levels in tissues sampled at different times); neither line of investigation has received attention in the past.

Ra-226 is biologically important due to its half-life (~1,622 y) and characteristic 4.78 MeV α–particle decay (Molinari and Snodgrass 1990). Its movement is of interest to radioecologists as it behaves environmentally and physiologically in ways similar to calcium (Ca^{++}) (Cowart and Burnett 1994). Concentration of ^{226}Ra in soft tissue does occur (Swanson 1983; Ruttenber et al. 1984), but at lower levels than in bone. Ra-226 is a bone-seeker that becomes incorporated in the bone matrix in the same way as its calcium analogue (Raabe et al. 1983).

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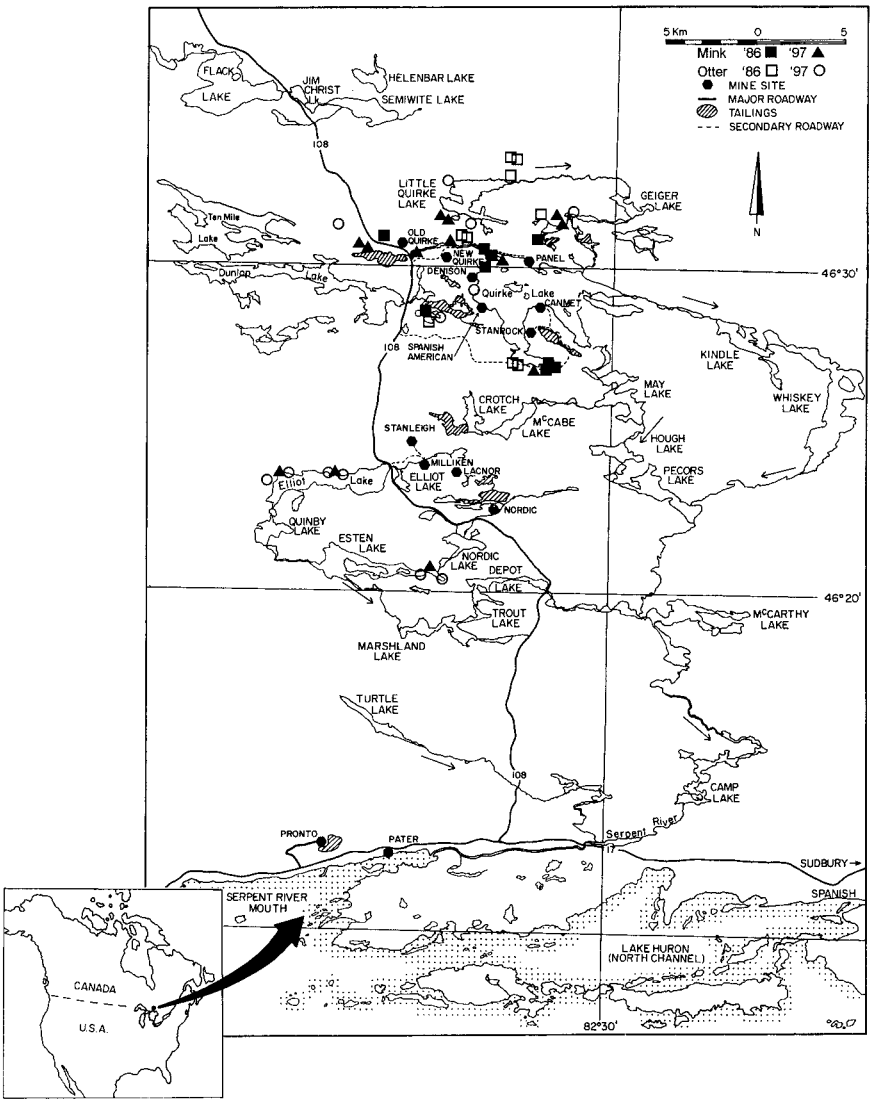

Figure 1. Mink and otter collection locations (1986 and 1997) in study area.

Two 250 mL aliquots of the stored liquid were analyzed for ^{226}Ra by the modified α–spectroscopic technique of Lim and Davé (1981). The method involves the addition of a ^{133}Ba working solution to the solid sample prior to ashing and chemical separation of radium by co-precipitation of Ba-Ra sulphates. Addition of the ^{133}Barium solution, with known activity, as a tracer, and subsequent measurement of the 356 keV γ–decay peak of ^{133}Ba provided a recovery factor (observed γ–count / expected γ–count) used to correct counts of the 4.78 MeV α–decay peak of ^{226}Ra. The final Ba-Ra precipitate was vacuum filtered on a Millipore HABP02500 (0.25 μm) filter which was then secured with double-sided tape to an aluminium planchette and counted for α–activity as described by Lim and Davé (1981). Blanks were included in each sample run to control for ^{226}Ra loss or contamination through the digestion/assay procedures. Recovery rates were assessed through the use of certified reference material (CANMET 'BL5'), obtained from the Mineral Science Laboratory, Canada Centre for Mineral and Energy Technology, Energy Mines and Resources Canada, and bovine shank samples spiked with and without a certified standard solution of ^{226}Ra. The precision and accuracy of the ^{226}Ra analytical method at the measured levels was better than 10% and the recovery rate of the spiked bovine samples was typically 98±10%. Instrument calibration was done as outlined by Lim and Davé (1981).

The simple linear concentration factor model (ICRP, 1978) was employed in this study. Comparison of the ^{226}Ra levels in the bone of the mink and otter, to published ^{226}Ra levels of some of their dietary items, allowed calculation of concentration ratios (CRs) useful as indications of ^{226}Ra movement through the ecosystem. As specified by ICRP, CRs were expressed on a fresh weight basis; when necessary, fresh–weight based data were estimated from dry–weight based data using standard conversion factors.

Between site and collection year comparisons were made on log-transformed data using one-way ANOVA, and the differences identified by Duncan's multiple range tests. Between species comparison of bone ^{226}Ra levels was made using the t-test for equal variances – a condition pre-confirmed using the Fisher test. Log–transformed ^{226}Ra data in otter bone reported by Wren et al. (1987) and those reported here had unequal variances (Fisher test). Accordingly, the two samples were compared using the t-test for unequal variances. For statistical computations, values reported below the limit of sensitivity (0.1 pCi g^{-1}) (Wren et al. 1987) were assigned a level of 3.7 mBq g^{-1}.

RESULTS AND DISCUSSION

Ra-226 levels in bone of mink and otter taken around Elliot Lake tailings (1986 and 1997) and from the distant reference areas (1997) are reported in Table 1.

Table 1. Ra-226 levels (mBq g^{-1} d.wt.) in bone of mink and otter taken near Elliot Lake uranium tailings in 1986 and 1997, and from reference areas.

Animal	Site	Date	n	Mean ± 1 SEM
Mink	Study	1986	9	38.2 ± 11.0
	Study	1997	13	28.8 ± 6.1
	Study	**pooled**	**22**	**32.7 ± 5.7**
	Reference	**1997**	**10**	**6.6 ± 2.2**
Otter	Study	1986	9	36.6 ± 9.4
	Study	1997	12	29.9 ± 10.7
	Study	**pooled**	**21**	**32.8 ± 7.2**
	Reference	**1997**	**10**	**3.1 ± 0.3**

Mean ^{226}Ra levels in bone of Elliot Lake mink did not differ significantly between years but were significantly higher ($F = 3.327$, $p < 0.001$) than that of the reference group. Similarly, there was no significant difference between ^{226}Ra levels of Elliot Lake otter bone taken in the two years, but both were significantly higher than in the reference group ($F = 3.34$, $p < 0.001$).

Interspecies comparison, using pooled data, indicated that bone ^{226}Ra levels in the Elliot Lake mink and otter were not significantly different ($t = -0.24$, $p = 0.41$). There was no significant difference ($t = -0.164$, $p = 0.43$) between our bone ^{226}Ra level in Elliot Lake otter (32.8 ± 7.2 mBq g^{-1} d.wt.) and that reported in bone of otters from the same area by Wren et al. (1987): 81.9 ± 64.3 mBq g^{-1} d.wt., recalculated from values expressed in $pCi.g^{-1}$, and assuming samples with ^{226}Ra levels below Wren's limit of sensitivity (0.1 pCi g^{-1}) had 3.7 mBq g^{-1} d.wt.

The similarity in mean ^{226}Ra values reported here for mink and otter bone, 32.7 and 32.8 mBq g^{-1} dry–weight respectively (Table 1), is not surprising as the feeding habits of the two animals overlap with common prey found in each diet. Mink are opportunistic predatory secondary consumers that are well adapted to hunting both aquatic and terrestrial prey; its dietary items include crayfish, fish, amphibians, and birds (Linscombe et al. 1982). The otter, also a secondary consumer, takes a variety of items – but with fish predominating in the diet (to 95%) (Toweill and Tabor 1982). The diets of both mink and otter are seasonal and depend on population levels of prey species (Linscombe et al. 1982;Toweill and Tabor 1982).

Bone ^{226}Ra levels reported here are less than those reported for the herbivorous (primary consumer) beaver (112.7 mBq g^{-1} dry–weight, Clulow et al. 1991) and muskrat (468.0 mBq g^{-1} dry–weight, Mirka et al. 1996) taken from contaminated waters in the same region. These herbivores consume aquatic roots and tubers, in the case of muskrat (Perry 1982), almost certainly with adherent mud and silt particles high in radionuclides, or vegetation harvested from the bank (and also at risk of being contaminated by radionuclide-rich particulates during spring runoff and flooding) and cached in lodges, in the case of beaver (Hill 1982). The lower levels in the carnivores can be explained by a limited intake of bone (the primary site of ^{226}Ra deposition) in relation to other tissues taken from prey animals.

Concentration ratios of ^{226}Ra to mink and otter bone from local lake water and from tissues of prey species of fish and other vertebrates of the study area (as reported in the literature) were calculated. Ra-226 levels (total) in lakewaters taken in the vicinity of Elliot Lake were reported at about 56 mBq L^{-1} (Clulow et al. 1998) yielding a calculated CR to bone (fresh-weight based) of the two species of 583; this falls in the upper part of the range of the water to bone CRs (30 – 650) reported by Swanson (1985). Concentration of ^{226}Ra from whitefish, lake herring, and laketrout bone to bone of both mammals ranged from 1.4 to 5.4 (using approximate levels in bones of specimens of the three fish species taken in the area of 23, 11, and 6 mBq g^{-1} respectively – Clulow et al. 1998). The corresponding CR values from muscle of area fish (containing on average from 0.4 to 0.6 mBq g^{-1}) to carnivore bone were 57 – 90 (same source as for bone). Higher CR values reflect the lower ^{226}Ra accumulation in prey species soft tissues than in bone (mentioned previously).

The feeding habits of both mink and otter are not limited to fish species but include other prey (Linscombe et al. 1982; Toweill and Tabor 1982). Mink and otter bone levels compared to reported muscle tissue levels in beaver (Clulow et al. 1991) indicate a CR of ~21. Calculated CR values to mink and otter bone from bone of the prey mammals beaver and muskrat (Clulow et al. 1991; Mirka et al. 1996), and the prey bird ruffed grouse (Clulow et al. 1992), were unity or less.

The lack of significant decline in bone ^{226}Ra levels in either species during the study period (during which U operations wound down then ceased) indicates inertia in the ecosystem. Clearly, change in radionuclide content of food items or water has not yet been of sufficient magnitude, or of sufficient duration, to be reflected in the tissues of the two secondary consumers examined.

Further understanding of the movement of radionuclides through the ecosystem in the area of U workings at Elliot Lake will require an examination of all food items of the mink and otter including other mammals, reptiles, amphibians, and aquatic invertebrates. Such study, in turn, could provide a greater empirical estimation and

understanding of relationships within various foodchains, using the sensitivity and precision associated with radionuclide measurements. In addition, the fate of a once-industrialized ecosystem may provide pointers for planning, operating, and closing of resource extraction and refining operations in the future.

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