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Uptake and Translocation of Naturally-occurring Radionuclides of the Uranium Series

D. C. Mahon

Department of Biological Sciences, Simon Fraser University, Burnaby, B.C., Canada, V5A 1S6

The mining of uranium ore and the subsequent construction of ore tailings piles releases radioactivity into the environment. Once released these radioactive materials are more readily available for uptake by animals and plants. To determine the incremental increase in the availability of radioactive materials it is necessary to know the potential for bioconcentration of radionuclides by the biota, the distribution of the radionuclides and the transfer coefficients for each radionuclide in each species in the food chain (BRENCHLEY et al. 1977). This information is generally site and species specific. Typically the amount of radioactivity in the various body compartments of each animal reflects the food source, behaviour and habitat of the animal (OSBURN 1974). TSIVOGLOU et al. (1960) described the accumulation of U_{nat} series radionuclides, U_{nat} and ^{226}Ra, derived from uranium mine tailings in the sediment, algae and fish of the Animas river system. However, very little work has been published on the uptake and accumulation of naturally occurring radionuclides from undeveloped ore bodies. The objectives of this paper are to delineate the uptake and transport of naturally occuring radionuclides of the U_{nat} series through three food chains in an area of high natural radioactivity, and to determine the potential for bioaccumulation of selected radioisotopes in the fauna.

MATERIALS AND METHODS

Water, vegetation and faunal samples were collected from an area of high natural radioactivity in the Okanagan Highlands of Central British Columbia (Canada). Mixed plankton samples were used in estimating radionuclide uptake; dominant zooplankter species were *Diaptomus sp.*, *Macrocyclops albidus* and *Daphnia rosea*. Dominant phytoplankters were *Melosira* and unidentified *Chlorophyta*. Benthos samples were collected by Ekman grab, hand sorted, and *Pisidium sp.* were analysed as a representative species. Water samples were acidified to 1% HNO_3 at the time of collection; subsequently they were filtered through a 0.3 um filter and digested with perchloric acid prior to analysis. Vegetation samples were collected by hand and frozen at -20C after collection. All vegetation samples were ground in a Wiley mill to pass through a 10 mesh. A portion of this material was dry ashed at 550C, digested in fuming perchloric acid and analysed for U_{nat}, ^{210}Pb and ^{226}Ra. A second portion of the material was wet digested in fuming perchloric acid and analysed for ^{210}Po. All whole animal samples were washed in distilled water, air dried at 110C for 48 h prior to analysis. Animal material was homogenised, ashed at 550C and digested as

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above prior to analysis for U_{nat}, ^{210}Pb and ^{226}Ra. Material for ^{210}Po analysis was treated as above. Uranium was measured by fluoresence on a Turner Model 111 fluorimeter, with a detection limit of 0.05 ppb (A.S.T.M. 1973). ^{226}Ra was separated by coprecipitation with barium sulphate. The radium-barium sulphate was stored for one week to allow ingrowth of ^{222}Ra and daughters. The precipitate was analysed on a Canberra Model 220 alpha- beta analyser (A.P.H.A. 1975). ^{210}Pb was measured indirectly by measurement of the beta activity of the daughter ^{210}Bi, after allowing 30 days for equilibrium. ^{210}Bi was extracted and counted as for ^{226}Ra, with a counting time of 100 min and a detection limit of 1 pCi/L. ^{210}Po was measured after electrodeposition on silver discs and alpha counting for 100 min, with a detection limit of 0.2 pCi/L (DOUGLAS 1967).

RESULTS AND DISCUSSION

Three foodchains were selected for examination, based on the fauna and flora of the region, information on potential accumulators of heavy metals or radionuclides and potential to reach man as the final acceptor in the chain. The following chains were examined, a) water sediment-algae-plankton-benthos-fish; b)browse vegetation-deer; c)vegetation-herbivorus mammals insectivorous mammals-carnivorous birds. In some instances the sample numbers available preclude statistical anaylsis of the data.

In the aquatic foodchain algae exhibit the highest concentration and bioaccumulation of radionuclides with the exception of ^{226}Ra. These data are in agreement with TSIVOGLOU et al. (1960) and ANDERSON (1961). It has been suggested that the apparent high bioaccumulation of radionuclides by algae may be due to surface adsorption of material rather than internal accumulation (ATKIN 1977). Plankton and benthos had comparatively high levels of U_{nat} and ^{226}Ra (Table 1). Both are known heavy metal accumulators (EDGINGTON et al. 1976; BROWN & ANASHULLAH 1971).

Table 1: Radionuclide concentration of media associated with the aquatic foodchain. (Sample size), Means$\pm$Std. dev.

	U-nat ppb	^{226}Ra pCi/kg	^{210}Pb pCi/kg	^{210}Po pCi/kg
Water (77)	0.34$\pm$0.17	<0.02	<0.5	0.217
Sediment(21)	10320$\pm$6180	2150$\pm$1130	4500$\pm$3690	3000$\pm$2500
Algae (1)	13400	20	2480	2430
Plankton(4)	600$\pm$210	340$\pm$210	NSS	NSS
Pisidium sp.(2)	400	300	NSS	NSS
S. gairdneri(17)	<5	<2	<20	12$\pm$10
(B)	40	15$\pm$6	75$\pm$34	30$\pm$20
C. catastomus(1)(F)	<5	<2	38	180
(B)	NSS	20	NSS	NSS

All values are for dry weight except water, concentration in water ppm or pCi/L.
F = flesh; B = bone, NSS=not sufficient sample for analysis.

Both fish species sampled, finescale suckers (Catastomus catastomus) and rainbow trout (Salmo gairdneri) contained low concentrations of U_{nat} and ^{226}Ra in both flesh and bone (Table 1). Finescale suckers had a higher burden of ^{210}Pb and ^{210}Po in flesh than rainbow trout. This probably reflects the bottom feeding habit of the suckers, and intake of sediment with high concentrations of both radionuclides (Table 1). Uranium levels in brown trout from uncontaminated streams in a uranium mining region of Czechoslovakia contained 10-40 ppb, but upon contamination of the streams from uranium mining these levels rose to 1600-2100 ppb within 3 years (JUSTYN & LUSK 1976). ^{226}Ra levels in uncontaminated fish from the same study were in the range 0.026-0.08 pCi/g whereas fish from contaminated streams contained 0.066-0.28 pCi/g. Trout from uncontaminated streams in uranium mining regions in Utah (USA) contained 0.001 pCi/g ^{226}Ra in flesh and 0.05 pCi/g ^{226}Ra in bone (SIGLER et al. 1966). Trout from a heavily contaminated uranium mining area in Canada (Elliot Lake, Ont.) contained 0.08 pCi/g ^{226}Ra in flesh (MACLAREN 1978). By comparison trout and suckers from the Okanagan Highland region, with 0.002 pCi/g ^{226}Ra in flesh and 0.015-0.020 pCi/g in bone can be considered uncontaminated (Table 1).

Table 2: Bioconcentration coefficients, water to biota. (CFx)[a/]

	U-nat	^{226}Ra	^{210}Pb	^{210}Po
Algae b/	1576	40	198	448
Plankton c/	459	4420	-	-
Pisidium (Mollusca)c/	306	3900	-	-
S. gairdneri (F)	14.7	<100	<40	55
(B)	118	750	150	138
C. catactomus (F)	14.7	100	76	830
(B)	-	1000	-	-

a/ $pCi.kg^{-1}$ tissue wet weight/ $pCi.L^{-1}$; or ppm U wet weight/ppmU
b/ Algal dry weight taken as 0.04 x wet weight (TSIVOGLOU 1960).
c/ Dry weight taken as 0.26 x wet weight (TSIVOGLOU 1960).

Calculated bioconcentration coefficients for ^{210}Pb and ^{210}Po in the organisms sampled compare well with those in the literature (Tables 2,3). The bioconcentration coefficients for U_{nat} and ^{226}Ra in benthos algae and plankton differ from those in the literature. However, ISKRA et al. (1970) report concentration factors for U_{nat} and ^{226}Ra, in algae, of about 100.

Table 3: Bioconcentration coefficients for edible aquatic organisms (THOMPSON et al. 1972).

	U-nat	^{226}Ra	^{210}Pb	^{210}Po
Algae	-	2500	200	2000
Crustacea	60	250	100	20000
Mollusca	60	250	100	20000
Fish	2	50	100	500

The data reflect the site specific nature of bioconcentration coefficient determination. In general the data uphold the contention that at each step in the foodchain there is a drop of about one order of magnitude in the bioconcentration coefficient relative to water, in agreement with BLAYLOCK & WITHERSPOON (1976), KOVALSKY et al. (1967) and THOMPSON et al. (1972), all of whom show U_{nat} and ^{226}Ra concentrations decreasing with increasing trophic levels; algae>invertebrates>fish.

In the terrestrial food chain (grass-forbs-lichen)-deer the composite lichen samples (Bryoria freemontia and Alectoria sarmentosa) contained the highest levels of ^{210}Pb and ^{210}Po but lower levels of ^{226}Ra. The grass, Calamagrostis rubescens, contained the highest levels of U_{nat}; and grouseberry, Vaccinium scoparium, contained intermediate levels of all four radionuclides (Table 4). Mule deer (Odocoileus hemionus), feed on browse lichen in winter, grass in spring and forbs in late spring and summer (SADLIER per. comm.) The deer showed differential sequesteration of the radionuclides to different body compartments (Table 4). Flesh contained the highest levels of U_{nat}, bone contained the highest levels of the other radionuclides, ^{226}Ra, ^{210}Pb and ^{210}Po. Liver tissue contained a high level of ^{210}Po compared to flesh.

Table 4: Radionuclide concentrations in the Deer foodchain. (Sample size), Means$\pm$Std. dev.

		U-nat ppb	^{226}Ra pCi/g	^{210}Pb pCi/g	^{210}Po pCi/g
C. rubescens	(7)	69.3$\pm$71.9	0.23$\pm$0.15	0.93$\pm$0.45	0.45$\pm$0.30
V. scoparium	(9)	16.2$\pm$10.4	0.29$\pm$0.39	1.84$\pm$0.63	2.28$\pm$0.80
Bryoria/Alectoria	(4)	17.5$\pm$10.4	0.05$\pm$0.03	8.98$\pm$1.91	9.89$\pm$1.97
O. hemionus	(2) (F)	24	0.005$\pm$0.002	0.028$\pm$0.009	0.04$\pm$0.06
	(B)	9	0.019$\pm$0.006	0.57$\pm$0.08	0.39$\pm$0.07
	(L)	5	0.004	0.03$\pm$0.01	0.12$\pm$0.02

F=Flesh; B=Bone; L=Liver

The large differences in the tissue content of ^{210}Pb and ^{210}Po compared to ^{226}Ra indicate the non-decay source of these radionuclides. If the ^{210}Pb and ^{210}Po were derived only from decay of ^{226}Ra they would be in equilibrium with ^{226}Ra, the unsupported nature of the daughter radionuclides results from their selective accumulation. Estimates of ^{210}Pb and ^{210}Po in the deer tissues are similar to those reported for reindeer in Alaska and Finland (UNSCEAR 1977). Reindeer meat is considered to be a critical pathway for human assimilation of these two radionuclides. The ^{226}Ra content of the flesh and bone of the deer was an order of magnitude lower than similar samples from the Elliot Lake area of Ontario (Canada), an area of high uranium waste pollution, whereas the ^{210}Pb and ^{210}Po levels in bone were significantly higher. It may be that the undisturbed nature of the ore deposit inhibits translocation of uranium and radium, but that exhalation of ^{222}Ra the parent radionuclide for ^{210}Pb and ^{210}Po, permits the accumulation of the daughter radionuclides in browse

vegetation. The data could indicate that grass would be the major source of U_{nat}, to deer, and that arboreal lichen could be considered the prime source for both ^{210}Pb and ^{210}Po, for deer.

Several small mammals, their preferred food species, and a predator were collected as examples of a carnivore foodchain (Table 5). The ^{210}Pb and ^{210}Po content of the small mammals was about an order of magnitude greater than the ^{226}Ra burden, and is in agreement with data from HOLTZMANN (1966).

Table 5: Radionuclide concentrations in members of the vegetation-small mammal-carnivore chain. Means$\pm$Std.dev.

		U-nat ppb	^{226}Ra pCi/g	^{210}Pb pCi/g	^{210}Po pCi/g
E. angustifolium		22.1$\pm$8.1	0.18$\pm$0.17	0.44$\pm$0.16	0.23$\pm$0.03
V. scoparium		16.2$\pm$10.4	0.29$\pm$0.39	1.84$\pm$0.63	2.28$\pm$0.80
T. hudsonicus	(F)	<4	0.007	0.06	NSS
	(B)	<50	0.05	0.18	NSS
E. amoenus	(C)	10	0.004$\pm$0.002	0.06$\pm$0.01	0.09$\pm$0.02
P. maniculatus	(C)	<5	0.013$\pm$0.005	0.036$\pm$0.008	0.13$\pm$0.02
M. pennsylvanicus	(C)	NSS	<0.004	0.05$\pm$0.03	NSS
L. americanus	(F)	<10	0.008$\pm$0.006	0.034$\pm$0.007	0.03$\pm$0.02
	(B)	<10	<0.008	0.09$\pm$0.04	0.06$\pm$0.03
C. corvax	(F)	NSS	0.003	0.003	NSS
	(B)	<5	0.044	0.26	NSS

All measurements on a dry weight basis. F=Flesh, B=Bone, C=Composite sample, NSS=not sufficient sample for analysis.

Deermice (Peromyscus maniculatus) and chipmunks (Eutamius amoenus) were observed feeding on fireweed seed heads (Epilobium angustifolia) and grouseberry. Grouseberry contained higher levels of both ^{210}Pb and ^{210}Po than other vegetation species in the area except lichen. Biomagnification of radionuclide content could not be detected in the raven (Corvus corvax), though this would have been expected from information on the accumulation of heavy metals by carnivorous birds (FIMREITE 1974).

The data suggest that the routes of uptake of ^{210}Pb and ^{210}Po in the terrestrial system are through the aerial parts of plants, as suggested by HILL (1967), but that neither U_{nat} nor ^{226}Ra is acccumulated to any great extent by terrestrial herbivores in areas of high natural radioactivity. Movement of naturally occurring radionuclides from undisturbed ore bodies into aquatic systems does not appear to result in high levels of contamination in fish. Sediments do accumulate high concentrations of radionuclides, as do the algal, planktonic and benthic populations. The levels of radionuclides in terrestrial animals inhabiting areas close to naturally occurring radionuclide sources do differ from those of animals and plants inhabiting areas near uranium tailings. There is a change in the ratio of ^{226}Ra:^{210}Pb and ^{210}Po in the body burden. There is consistancy in the rule that for each trophic level there is a drop of one order of magnitude in the body burden

of the four radionuclides considered. The data indicate that specific components of the environment; sediments, algae, lichen, and grass are major sources for the body burden of specific radionuclides in each food chain. The aquatic system is the most sensitive to pertubation from accumulation of radionuclides because of the presence of a variety of accumulator mechanisms, both biotic and abiotic. The critical sequence in the foodchain is the algae plankton step. Algae would be a suitable indicator species for the detection and measurement of aquatic contamination by radionuclides.

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