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RADIONUCLIDE LEVELS AND ^{226}Ra CONCENTRATION RATIOS BETWEEN WATER, VEGETATION, AND TISSUES OF RUFFED GROUSE (*Bonasa umbellus*) FROM A WATERSHED WITH U TAILINGS NEAR ELLIOT LAKE, CANADA

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RADIONUCLIDE LEVELS AND ^{226}Ra CONCENTRATION RATIOS BETWEEN WATER, VEGETATION, AND TISSUES OF RUFFED GROUSE (Bonasa umbellus) FROM A WATERSHED WITH U TAILINGS NEAR ELLIOT LAKE, CANADA

F.V. Clulow*, T.P. Lim** and N.K. Dave+

ABSTRACT

Radionuclide levels measured in bone, muscle, kidney, and liver tissues, gut contents, and diet items of 47 grouse from the Serpent River drainage basin, containing U tailings at Elliot Lake, and from control areas in Ontario, showed variation by site and tissue.

The mean level of ^{226}Ra in bones of grouse sampled from Elliot Lake (28.5 $mBq.g^{-1}$) was higher than that in bones of birds from a distant control site near Sudbury, Ontario (8.0 $mBq.g^{-1}$), but similar to the value in a local control population (28.1 $mBq.g^{-1}$). Birds from Mid- and Low-Serpent River basin populations (with 17.1 and 17.7 $mBq.g^{-1}$ respectively) did not differ from local or distant control populations; muscle, liver, and kidney had lower ^{226}Ra concentrations which did not differ significantly among populations. Levels of ^{226}Ra in the crop contents and chyle did not differ significantly by site and were similar to those of food items consumed by the birds. Chyme values were higher in birds sampled in Elliot Lake and at the local control site than in those taken at the distant control site; birds sampled downstream from Elliot Lake did not differ from distant controls in this regard.

Levels of ^{232}Th, ^{230}Th were below detection limits (0.1 $\mu g.g^{-1}$ and 5.0 $mBq.g^{-1}$ respectively) in bone, muscle, and liver tissue in two grouse with elevated levels of ^{226}Ra. Other radionuclides were measurable in some tissues: ^{238}U in bone at 0.4 $\mu g.g^{-1}$, in muscle to 0.2 $\mu g.g^{-1}$, in liver to 1.0 $\mu g.g^{-1}$; ^{228}Th was found only in muscle (8.0 $mBq.g^{-1}$); ^{210}Po was found in bone, muscle, and liver (maxima: 24.0, 7.0, 16.0 $mBq.g^{-1}$) with the exception of one muscle sample; ^{210}Pb was detected in only one liver sample (50.0 $mBq.g^{-1}$).

Key words: Uranium tailings; Ruffed grouse; Radionuclides uptake; ^{226}Ra.

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Environmental levels fell within ranges previously reported at the sites, or at similar locations elsewhere. Leaves of trembling and largetooth aspen growing in the basin had mean ^{226}Ra levels of 41.8 and 52.7 $mBq.g^{-1}$ (dry weight) respectively, fungal material carried up to 215.4 $mBq.g^{-1}$ (air-dried), with some variation by site. River and lake waters sampled near the U tailings had 118.1 $mBq.L^{-1}$ of dissolved ^{226}Ra and at the distant control site the value was 12.1 $mBq.L^{-1}$.

The concentration ratios (CR) between bone of grouse collected at the Elliot Lake sites and trembling and largetooth aspen leaves were 1.38 and 1.09 (fresh weight basis); from other diet items and other tissues the values were less than unity. Water to tissue bone ratios, based on dissolved ^{226}Ra levels ($mBq.L^{-3}$) ranged to 144.08.

People eating grouse from the study area are unlikely to consume radionuclides in excess of limits currently established by Canadian regulatory authorities.

INTRODUCTION

The work reported here forms part of an investigation into the movements of radionuclides through components of the Serpent River drainage basin ecosystem which contains deposits from thirty years of U mining and milling operations at Elliot Lake, Ontario (Figure 1). The purpose of the study was to obtain information on ^{226}Ra and other radionuclide levels for use in calculation of concentration factors useful in modelling contaminant movement and to permit estimation of radionuclide intake by human consumers of wild game from the area. Samples from control sites were taken to provide background values of animals, vegetation, and water not associated in any way with U wastes.

To achieve the purpose of this study, ^{226}Ra levels were measured in: a) bone, muscle, liver, and kidney tissues of ruffed grouse (Bonasa umbellus) taken in the Serpent River basin, and from control sites; b) vegetation items consumed by the animals, and whenever possible, their gut contents; and c) water used by the animals for drinking. Other radionuclides (^{238}U, ^{232}Th, ^{230}Th, ^{228}Th;, ^{210}Pb, and ^{210}Po) were measured in tissues of two grouse with elevated tissue levels of ^{226}Ra.

Effective tailings management and rehabilitation requires understanding and knowledge of radionuclide movement through the ecosystem of which the tailing deposits form a part. Modelling is commonly used to assist this understanding and to predict behaviour of the contaminated ecosystem. In turn, modelling requires knowledge of transfer parameters between biologically linked components of the system (e.g., substrate and plant tissue; plant and herbivore). The concentration ratio (CR) is a transfer parameter defined as the ratio between steady-state concentrations in connected compartments of a model (ICRP 1978) such as an animal's diet (vegetation) and the animal's tissue (e.g., bone). The CR, while not an indicator of the effects of radionuclide concentration on an organism, is a tool by which differences in accumulation by organisms may be assessed and is useful in modelling equilibrium conditions.

Information on radionuclide movements in the progression substrate-vegetation-animal (tissue) is limited. Food and tissues of small terrestrial mammals (voles, Microtus pennsylvanicus) and larger game animals (deer, Odocoileus virginianus, moose Alces alces) from the vicinity of the U tailings, or the drainage from these tailings, at Elliot Lake, Ontario, have been studied under wild and natural conditions to provide data useful in this regard (Burns et al. 1986; Cloutier et al. 1983, 1985a,b, and 1986; MacLaren 1978, 1987). Ra-226 levels in faeces of hares (Lepus americanus) from the area have been reported (Clulow et al. 1986) as have levels in cutworms (Agrotis ipsilon) eaten by gulls (Larus argentatus) visiting the tailings (Clulow et al. 1988). Clulow et al. (1988X) and Mirka et al. (1988X), have reported on beaver and muskrat levels. Information on radionuclide levels in food or tissues of game birds of the area is limited: levels of ^{226}Ra in muscle tissue of three ducks and two grouse were given by MacLaren (1978).

Knowledge of the food habits and movements of ruffed grouse was taken into account in the design of the study and subsequent interpretation of results. Ruffed grouse consume plant species in proportions varying with location and season and do not take appreciable amounts of animal material in their diets. There is general consistency in the literature suggesting that northern grouse favour aspen* leaves and buds over those of all other trees (Svoboda and Gullion 1972; Doerr et al. 1974), although willow is also taken

*Common names of plants after Anonymous, 1961.

when available. Rose and Parker (1983), reported Northern Ontario grouse had much aspen material in their crops at two sites in the two seasons studied. Other food items, including arthropods, horsetails, and fungal tissue accounted for a substantial proportion of the gut content (>50% dry weight and >60% by frequency of crop contents in the case of fungi) of birds from one collection site.

It was anticipated that levels of ^{226}Ra in the contents of the alimentary canal of the birds studied would reflect levels in the food items consumed and that levels of chyme (stomach contents) might be higher than in samples taken from other parts of the gut because birds pick up and retain grit (possibly rich in contaminants) for use as grinding stones in the gizzard.

Movements of grouse, studied by banding nestlings and subsequent recovery of them when older, start in the fall, when family groups disperse. Exceptionally, dispersal movement up to 19.2 km has been reported, but 72% of marked birds travel less than 1.6 km from the point of banding (Hale and Dorney 1963). Adult grouse are more sedentary than younger birds, and females are more mobile than males (Archibald 1976; Godfrey 1975; Chambers and Sharp 1959; Hale and Dorney 1963; and Gullion and Marshall 1968). Birds reported on in this study may have moved during their lives and were not necessarily long-term residents at their place of capture.

METHODS AND MATERIALS

SAMPLING LOCATIONS

All samples came from the Sudbury-North Bay and Timagami sections of the Great Lakes -St. Lawrence Forest Region of Canada (Rowe 1959) described in detail elsewhere (Clulow et al. 1988X). This region has rugged outcrops of Canadian Shield bedrock among wet flats and lowlands variably covered with mixed hardwoods and conifers.

Study areas (Figure 1) in the Serpent River drainage were three in number: a) 'Elliot Lake', a triangular area with points at Ten Mile Lake to the Northwest, Rochester Lake in the Northeast, and Nordic Lake to the South, which contains the Town of Elliot Lake, U tailings deposits, and several lakes and waterways. Substantial variation in ^{226}Ra concentration in these water bodies has been reported (see below). b) 'Mid-Serpent', the watercourse downstream from the river's exit from Quirke Lake, contains Bear, Whiskey and Pecors Lakes. Land adjacent to this stretch of the river, unpopulated except for fishing camps, contains several registered traplines. c) 'Low-Serpent', the lower reach of the river, below its exit from Pecors Lake, passes through the territory of the Serpent River Indian Band, whose members trap along it, before emptying into the North Channel of Lake Huron.

Control areas were at two locations: a) 'Control-distant', 40 km NE of Sudbury and more than 130 km E of Elliot Lake, contains Lakes Wanapitei, Boot, Rathbun, Portage, and Matagamasi, and was chosen for its remoteness from Elliot Lake and its ease of access. Several U occurrences have been reported in the area, but none have been developed (W. Meyer, Resident Geologist, Ministry of Northern Development and Mines, Government of Ontario, Sudbury; personal communication). b) The second 'Control-local', encompassing Tweedle, Sagard and Poulin Townships, located about 40 km NW of Elliot Lake, upwind and in a different watershed, was chosen because of accessibility, location, and lack of U industry operations. Occurrences high in Th-series radionuclides are known in this general area (W. Meyer; personal communication).

Water quality records of the Ministry of the Environment of the Province of Ontario (MOE) (MOE 1981, 1982, 1987) indicate substantial variation in ^{226}Ra concentration from place to place and over time at MOE sample stations in the vicinity of Elliot Lake.

ANIMAL SPECIMENS

Because tissue levels were expected to vary from animal to animal, a goal of ten or more grouse specimens was set as the desirable sample size (n) at each location studied. Measurements were made of ^{226}Ra levels in muscle, bone, kidney, liver, and gut content (when sufficient material was present) of each animal. As information was available on radionuclides in water and vegetation of the study areas, estimates were restricted to four water samples and four samples of plants used by the animals for food in each area. Estimations of total and dissolved ^{226}Ra in the water samples were made. Ra-226 levels in aliquots of vegetation samples (pooled by species) were measured.

Collecting - Grouse, purchased from hunters, were harvested from September 1987 through late winter 1988. Samples were labelled as received and their points of origin recorded.

Processing: Measurement, Dissection, and Tissue Separation - Carcasses, received frozen, were mostly intact except for several lacking the pectoralis (flight/breast) muscles, which had been removed for eating, internal organs due to evisceration, or legs discarded during dressing the birds. Specimens were thawed and weighed. Dissection, inspection and removal of internal organs, and sexing, followed. Samples of pectoralis or leg muscle tissue, the liver, both kidneys and the hind legs (for muscles and bone) were separately bagged and frozen for later study. Bones of the hind legs were prepared in a similar manner to that employed for beaver femurs (Clulow et al. 1988X). In specimens lacking legs, the sternum and furcula were analyzed in place of limb bones.

Aging - Grouse were aged as immature (I), aged under 1 y, and adult (A), aged 1 y or more, on the basis of replacement and growth of the primary and secondary flight feathers of the right wing (Hale et al. 1954). An animal is adult if primary flight feathers 8, 9 and 10 are rounded with sheathing at the base of the feather; juveniles show primary flight feather 8 rounded at the tip and sheathing is present at the base, but primaries 9 and 10 are pointed and lack sheathing (ruffed grouse only molt through primary 8 in their first fall).

PLANT SPECIMENS

Collecting - Woody plant material was collected as described by Clulow et al. (1988X). Fungi (mushrooms and toadstools) were collected in winter 1987-1988 from decaying tree trunks at: Elliot Lake (Stanrock Mine turnoff, 50 m east of Hwy 108) on 16 November; Mid-Serpent (Whiskey Lake;, east shore) on 1 October; and Low-Serpent (intersection of Hwy 17 and Serpent River) on 7 November. Control samples were taken on the eastern shore of Lake Wanapitei on 5 April. Fungal samples were brought frozen to the laboratory where they were identified and stored dry until analysis.

Processing - Woody plant material was processed as described by Clulow et al. (1988X).

WATER SAMPLES

Collection and processing of water samples has been described elsewhere (Clulow et al. 1988X).

ANALYSIS OF ^{226}Ra

Measurement of ^{226}Ra in duplicate solution samples was by the alpha-spectroscopic method of Lim and Dave (1981): the 4.78 MeV alpha-decay peak of ^{226}Ra was counted following precipitation of Ra-Ba sulphate. A ^{133}Ba tracer solution, added to starting solid samples, allowed measurement (and appropriate correction) for the overall analytical recovery for ^{226}Ra analysis. Recovery rates of 80% or more of the amount of ^{133}Ba added by spiking were usual in the study.

ANALYSIS OF OTHER RADIONUCLIDES

Duplicate 1 g (approx.) samples of dried bone, muscle, and liver from two grouse were sent for radionuclide measurement to the laboratories of Atomic Energy of Canada Limited (AECL), Kanata (neutron activation analysis) and Monenco Science and Technology (MONENCO), Calgary (chemical separation and alpha-spectroscopy). AECL reported results of neutron activation analyses of U-238 and ^{232}Th (delayed neutron counting) in ppm, with threshold values at 1 ppm. MONENCO reported values in $mBq.g^{-1}$ with threshold values at 5 $mBq.g^{-1}$ for ^{226}Ra, ^{232}Th, ^{230}Th, ^{228}Th and ^{210}Po, and 50 $mBq.g^{-1}$ for ^{210}Pb. In the case of ^{210}Pb, analysis was performed by measurement of its daughter, ^{210}Po, after 42 d in-growth to the samples.

QUALITY ASSURANCE

Several approaches assure the reliability of the ^{226}Ra data reported here. First, analyses were carried out in the laboratories of CANMET, Elliot Lake, where routine procedure involves calibration of the analytical system itself by certified NBS calibration standards. Second, the standard method, involving recovery of the gamma emitter (^{133}Ba) added to samples prior to digestions to measure analytic recovery of the radiological procedure, when applied to samples of cow shank bone (NBS material) spiked with known amounts of ^{226}Ra and ^{133}Ba, showed a recovery rate of 98 $\pm$ 10%. Third, the Quality Asssurance Program of the CANMET Laboratory has the following checks made as a matter of routine: a) Certified CANMET tailings samples DL-1 and DL-2 are analyzed; b) Standards and blanks are analyzed along with samples; c) Cross-checks on liquid, solid, and biological sample measurements by CANMET are carried out by MONENCO Analytical Laboratories (Calgary). These three checks indicated the variation was $\leq$10%.

As part of the current study, an additional check was carried out: two fragments of two grouse bones were analyzed independently (blind) by MONENCO

Analytical Laboratories of Calgary, and by CANMET (see Results section).

DATA

Statistics and Transformations - As it was suspected that the data were unlikely to be normally distributed, nonparametric tests were performed for initial examination of differences among groups of animals divided by sex and age. Mann-Whitney U tests (two-tailed) were used to compare pairs of sub-groups. Significance was indicated ($P < 0.05$) when $U_{calculated} > U_{table}$, (alpha = 0.05) (Sokal and Rohlf 1979). Kruskal-Wallis tests were employed for comparisons among three or more subgroups groups. Significance was indicated ($P < 0.05$) when $K_{calculated} > X^2_{table}$ (alpha = 0.05) (Lapin 1975).

Differences among site categories and areas were tested at the 5% level using a one-way analysis of variance (ANOVA) on log-transformed data. Fisher's protected LSD test (PLSD) was then performed on area and site groupings to see if significant differences occurred among Elliot Lake sites and Control areas.

Presentation and Calculations - To facilitate comparisons with the findings of other workers, values are expressed throughout as $mBq.g^{-1}$ dry weight of plant or animal tissue and $mBq.L^{-1}$ of water, except in the calculation of concentration ratios, where the use of values based on wet or fresh weight, and $mBq.g^{-1}$ of water, is called for by convention.

Graphical presentations are of back-transformed data. Sample means and their associated standard errors were anti-log transformed to ensure that units on the vertical axis are familiar; this process accounts for error bars appearing asymmetric in some cases.

CONCENTRATION RATIOS

When calculating the ^{226}Ra concentration ratio (f_{1e}), the following formula (after ICRP 1978) was employed:

$$f_{1e} = X_1/X_e$$

where X_1 = radionuclide concentration in animal tissue (mBq.g^{-1} wet weight)

and X_e = radionuclide concentration in water (mBq.g^{-1}) or vegetation (mBq.g^{-1} wet weight).

Mean animal tissue and gut content ^{226}Ra concentrations were calculated on log-transformed data and concentration ratios were based on their back-transformed values. For these calculations radionuclide concentrations were expressed as mBq.g^{-1} wet weight as required by the standard formula. Radionuclide concentration in diet items needed to be calculated as the level at the time of consumption. This presented a problem since moisture content of vegetation varies with time of day and season due to water stress (Wilson et al. 1953; Boyer 1968), and with changes resulting from absorption or loss of water after removal from the living tree. Tissue moisture is also lost in varying amounts due to differences in handling, and the time and method of sample storage prior to analysis.

To reduce variation attributable to variation in moisture content, values for vegetation samples, originally expressed as mBq.g^{-1} dry weight, were adjusted to standard moisture contents. In a study of moisture in leaves picked in the vicinity of Sudbury, Ontario, in September, G.M. Courtin of Laurentian University (personal communication) observed the moisture content of aspen leaves to be 69.1 $\pm$ 1.67%, n = 4, of their fresh weight. Leaves left in protective net bags on forest floor for 42 d absorbed water: 10.2 $\pm$ 8.39%, n = 3, of their fresh weight in the case of aspen. In calculating radionuclide levels in leaves used as diet items in the present study, a value of 70% moisture content was used as a standard value.

As fungal material is eaten by grouse after being dried out by wind and sun in winter, the radionuclide level in air-dried material was used as the basis for calculation of concentration ratios.

RESULTS AND DISCUSSION

A total of 47 ruffed grouse were collected in the winter 1987-1988: 9 from Elliot Lake, 7 from Mid-Serpent, 9 from Low-Serpent, 10 from Control-local, and 12 from Control-distant locations. Low population density of birds in Elliot Lake, Mid-Serpent, and Low-Serpent areas during the regular shooting season and the following winter caused the shortfall from the desired n $\geq$10 goal for these locations. Kill sites around the Elliot Lake tailings are indicated in Figure 1.

Levels of ^{226}Ra were measured in bone, muscle, liver, and kidney tissues of most grouse specimens collected, as well as in their crop contents, chyme, and chyle (contents of the intestines). Several Low-Serpent specimens were received incomplete or damaged and gut samples from these were not obtainable. Information on grouse tissue and gut content levels of ^{226}Ra is summarized in Figures 2 to 6.

Levels of ^{226}Ra in bone of grouse from each area are indicated in Figure 2. ANOVA indicated significant difference(s) among the groups ($F_{4,42}$ = 3.33 P <0.05). Fisher's PLSD test revealed the following differences among treatments:

		Elliot Lake	Control-local	Mid-Serpent	Low-Serpent	Control-distant
Mean [^{226}Ra] $mBq.g^{-1}$	=	28.5	28.1	17.7	17.1	9.6

(Back-transformed means presented, those sharing same underline are not significantly different, P $\geq$0.05.)

Levels of ^{226}Ra in muscles of birds taken in each area are indicated in Figure 3. As analysis of variance on the log-transformed data failed to indicate any significant difference among the groups ($F_{4,41}$ = 1.94, P $\geq$0.05), no further analyses were carried out. Concentrations of ^{226}Ra in liver samples from grouse taken at each site are seen in Figure 4. Analysis of variance on the log-transformed data failed to indicate any significant difference among the groups ($F_{4,37}$ = 0.73, P $\geq$0.05) and the data were not analyzed further. The levels of ^{226}Ra in kidney tissues taken from grouse sampled at each location are indicated in Figure 5. Analysis of variance on the log-transformed data failed to indicate any significant difference among the groups ($F_{4,39}$ = 0.83, P $\geq$0.05), so no further analyses were carried out.

Levels of ^{226}Ra in gut (crop, stomach, and intestine) contents of grouse taken from different areas are plotted in Figure 6. Analysis of variance on the log-transformed data indicated significant differences only among levels of stomach contents from birds taken at different locations ($F_{4,36}$ = 7.70, P <0.05). Fisher's PLSD test indicated the following:

	Elliot Lake	Control-local	Mid-Serpent	Low-Serpent	Control-distant
Mean [^{226}Ra] $mBq.g^{-1}$ =	22.2	18.42	15.42	9.62	6.98

(Back-transformed means presented, those sharing same underline are not significantly different, P $\geq$0.05.)

Crop and intestine content levels did not differ significantly by site ($F_{4,33}$ = 1.95, and $F_{4,37}$ = 0.0.42 respectively, P$\geq$0.05 in both cases). There was no significant variation among crop content, chyme, and chyle values at each site.

The mean level of ^{226}Ra in bones of grouse sampled from Elliot Lake was higher than that in bones of birds from the distant control site, but similar to the value in the local control population; grouse from lower in the Serpent River drainage basin are not distinguishable, on the basis of their bone ^{226}Ra burdens, from those of the local or distant control areas. Mean levels in muscle, liver, and kidney did not vary significantly by site. Ra-226 levels of 1.5 and 1.9 $mBq.g^{-1}$ dry weight of two grouse muscle samples, and

1.1 to 4.1 $mBq.g^{-1}$ dry weight in three duck muscle samples, were reported by Anonymous (1978a).

Levels of ^{226}Ra in the crop contents, chyme and chyle of birds sampled at Elliot Lake were similar to levels in food items measured. Crop content and chyle values did not differ significantly by site. Chyme values were higher in birds sampled in Elliot Lake and at the local control site than in those taken at the distant control site; birds sampled downstream from Elliot Lake did not differ from distant controls in this regard. Contrary to expectations, grouse chyme did not have higher levels of ^{226}Ra than samples taken from other parts of the gut. Although the birds pick up and retain pebbles and small stones for grinding food in the gizzard, they do not appear to select particles rich in ^{226}Ra. However, chyme of birds taken close to the tailings did have more ^{226}Ra than that of birds taken at the distant control site.

VEGETATION - WOODY PLANTS

Levels of ^{226}Ra measured in woody plant samples are seen in Table 1. The ^{226}Ra levels in trembling aspen, sampled from the vicinity of tailings in the Elliot Lake area, agree with data reported previously on samples taken from the tailings themselves: Dave et al. (1985a), reported trembling aspen from the tailings with 33 $\pm$ 7 to 126 $\pm$ 11 $mBq.g^{-1}$ dry weight in leaves; Kalin (1988) found mean ^{226}Ra values of 92.5 $mBq.g^{-1}$ in trembling aspen leaves from several sites on the tailings. Levels in plants at the Control-distant area generally agree with control values of Dave et al. (<4 $mBq.g^{-1}$ dry weight).

VEGETATION

Levels of ^{226}Ra measured in fungal tissue samples are seen in Table 2. Fungal tissue levels, similar among species, tended to decrease with distance from the tailings. There are no published data to which these values might be compared.

WATER

Levels of ^{226}Ra measured in water samples are seen in Table 3. Total ^{226}Ra concentrations reported here for water taken from the Serpent River, and the lakes through which it passes, are higher than in moose drinking water sampled in the region and at a control site (range 5-60 $mBq.L^{-1}$, MacLaren 1987). However, samples in that study were collected some distance from the Serpent River. Control-distant values are close to the limits of sensitivity of the analytic method employed and differences between total and filtered concentrations, and between our results and those for controls in studies in this area (5 $mBq.L^{-1}$, MacLaren 1987) and elsewhere (20 $mBq.L^{-1}$, Swanson 1985) are not considered important.

Total ^{226}Ra concentrations reported in Table 3 generally agree with others found in the Elliot Lake region. Most fall within the ranges reported by MOE for equivalent sites (see Table 7). The Elliot Lake sample, with 558.7 $mBq.L^{-1}$, corresponds to MOE site 014 which had a range of 80-610 $mBq.L^{-1}$, in the period from 1984 to 1987. The Mid-Serpent water, showing 192.0 $mBq.L^{-1}$, corresponds to MOE site 035 sampled once in 1984 and again in 1985 with a total ^{226}Ra concentration of 60 $mBq.L^{-1}$ found each time. However, water entering Whiskey Lake is the outflow of Quirke Lake with a range of 20-970 $mBq.L^{-1}$ from 1984 to 1987 as measured at MOE site 049. The Low-Serpent values of 232.0 and 358.7 $mBq.L^{-1}$ fall within the range for MOE site 001 of 30-600 $mBq.L^{-1}$.

Consonance of these data with others from the area, their general agreement with expectations for the area, and the decrease in ^{226}Ra concentrations with distance downstream from tailings, leads to confidence in the reported values.

OTHER RADIONUCLIDES

Levels of other radionuclides, as reported by AECL and MONENCO, in tissues of two grouse, are seen in Table 4; CANMET ^{226}Ra data have also been included for comparison.

Muscle levels of ^{238}U presented in the Table (<0.1 and 0.2 $\mu g.g^{-1}$ dry weight) compare to values previously reported in two grouse taken from the Elliot Lake area (<0.05 and 0.08 $\mu g.g^{-1}$ dry weight) (Anonymous 1978a). Levels of ^{232}Th, ^{230}Th, and ^{210}Pb were similarly below detection limits in all cases.

CONCENTRATION RATIOS

Concentration ratios were calculated between 'diet' items and tissue compartments of grouse taken from the site (Table 5).

Rose and Parker (1983) reported gut contents of grouse varied with season and site. In crops of animals collected in the fall, proportions of the contents (dry weight basis) were reported by them as follows: aspen leaves from 15-60%; fungi from 10-60%; and various other materials to 30% in aggregate. A typical diet, based on these observations, might have the following proportions: aspen leaves 45%, fungi 45%, others (including arthropods) 10%. Rose and Parker reported blueberry leaves among the other items in the diet of these birds; these plants also occur in the area of Elliot Lake, and Dave et al. 1985b, have reported total ^{226}Ra contamination of berries varying from 20 to 290 $mBq.g^{-1}$ dry weight within 500 m of tailings but declining to background levels of 2 to 6 $mBq.g^{-1}$ dry weight at distances of 1 km or more downwind from the tailings. About 17% of the contamination was removed by rinsing in distilled water. Taking a value midway between the extremes (155 $mBq.g^{-1}$ dry weight), removing 17% (26.4 $mBq.g^{-1}$ dry weight) to allow for adherent particulates, and converting the resulting value (128.7 $mBq.g^{-1}$ dry weight) to a fresh weight basis value (assuming a moisture content of 70%, Dave et al. 1985b) gives an estimate for fresh blueberries of 128.7x 30/100 = 38.6 $mBq.g^{-1}$ wet weight. If the berry content is a reflection of the ^{226}Ra content in the rest of the plant, and this is similar to the levels in other plants in the area,then we can use this value for the 10% of the diet made up of plant and other materials. Although higher than aspen leaves, the value falls within the range for plant values reported in Table 3 (adjusted for moisture content).

In a diet with the components in the ratios just stated (45:45:10), with ^{226}Ra values of 12.5, 26.5 and 38.6 $mBq.g^{-1}$ wet weight in washed aspen leaves, washed fungi, and washed other items, the weighted average level of ^{226}Ra is 21.4 $mBq.g^{-1}$ wet weight. This value compares to the value of fresh crop contents (12.36 $mBq.g^{-1}$ wet weight). When the vegetation value is considered with the mean tissue levels presented in Table 5, the following concentration ratios (wet weight basis) emerge:

Bone/vegetation	17.3/21.4	=	0.81
Muscle/vegetation	1.0/21.4	=	0.05
Liver/vegetation	3.2/21.4	=	0.15
Kidney/vegetation	7.8/21.4	=	0.34

Concentration ratios between crop content and grouse tissue levels are as follows (dry weight basis):

Bone/crop content	29.3/34.3	=	0.85
Muscle/crop content	3.8/34.3	=	0.11
Liver/crop content	9.8/34.3	=	0.29
Kidney/crop content	27.9/34.3	=	0.81

Concentration ratios from diet to bone are higher than those to other tissues; this probably reflects the radionuclide being incorporated in the bone matrix in the same way as its analog Ca (Friedlander and Kennedy 1962; Lloyd et al. 1976; Raabe et al. 1983). Concentration in soft tissue (Mahon 1982; Schlenker et al. 1982; Swanson 1983; Ruttenber et al. 1984) does occur, but to a lesser extent than in bone. Bound in calcium hydroxyapatite bone crystals, ^{226}Ra may cause tissue damage, possibly resulting in osteosarcoma, as it is an internal emitter of alpha and beta radiation (Van Dilla et al. 1958; Mays et al. 1975; Schlenker et al. 1982; Raabe et al. 1983).

Concentration ratios between diet items and bone reported here are similar to those reported for beaver (0.57 to 5.38) (Clulow et al. 1988X) and muskrat (1.07 to 8.47) (Mirka et al. 1988X) taken at Elliot Lake. In moose, CRs (calculated using 95% confidence intervals) from vegetation to bone were reported from 1.30 to 7.04 (MacLaren 1987). Wild meadow voles trapped in Elliot Lake are reported to eat diet items with a mean ^{226}Ra level of 211 $mBq.g^{-1}$ dry weight and have bone levels of 1.506 and 703 $mBq.g^{-1}$ dry weight in summer and fall; CRs calculated from these values are 7.1 and 3.3 (Cloutier et al. 1986). Burns et al. (1987), working with young meadow voles in the laboratory, reported a mean ^{226}Ra level of 59 $mBq.g^{-1}$ dry weight in bone of animals fed a diet spiked with ^{226}Ra (350 $mBq.g^{-1}$ dry weight); these values translate to a CR of 0.16 during the period of rapid bone growth studied. Muth and Glöbel (1983) calculated a concentration ratio of 23.7 between ^{226}Ra in food and human tissue.

The observation that grouse CRs from diet to soft tissues (muscle, liver, and kidney) are less than unity corresponds to observations made on the same tissues in beaver (Clulow et al. 1989X) and moose muscle and liver (MacLaren 1987).

HUMAN CONSUMPTION - ESTIMATES OF ANNUAL INTAKES

Grouse are considered a delicacy and are sought after by residents of the Serpent River basin who usually eat them roasted. Estimates of annual consumption involve assumptions regarding quantities of tissue consumed per year. Goldfarb (1977) reported local hunters consume 46 kg of game per family per year. This consumption supplements or replaces dietary animal material from other sources. According to Health and Welfare Canada (HWC 1975) average annual consumption of all meats by Canadian males 20 to 29 y old is 71 kg. To make a very conservative assessment, it is assumed here that a resident of the Serpent River basin might obtain all his animal material from wild sources and that muscle tissue, liver tissue, and bone particles are consumed in the (arbitrary) ratio of 100:10:1. These assumptions lead to an estimated daily consumption of about 175 g of muscle, 17.5 g of liver, and 1.8 g of bone.

The above tissue consumption rates, used in conjunction with the tissue radionuclide values of Table 4, permit estimation of annual intakes of radionuclides from grouse. Detection limit values were used in those cases in which a radionuclide was not measurable in a tissue sample from one animal, but was measured in the corresponding tissue in the second representative examined. Values were converted to $mBq.g^{-1}$ wet weight using the mean water contents of tissues measured during ^{226}Ra estimations (n = 10 in all cases): bone 41.5%, muscle 72.6%, liver 67.9%. For levels of ^{226}Ra in bone, both the CANMET and MONENCO values were included in the calculations; CANMET values were used for ^{226}Ra in muscle and liver.

Calculated annual intakes of radionuclides from grouse tissues are:

	Bone	Muscle	Liver	Total	% of Derived Limit on Annual Intake
^{238}U (mg)	<1	6	<1	<8	<0.12
^{232}Th (mg)	nm	nm	nm	-	-
^{232}Th (Bq)	nm	nm	nm	-	-
^{230}Th (Bq)	nm	nm	nm	-	-
^{228}Th (Bq)	nm	127	nm	127	0.25
^{210}Po (Bq)	9	117	353	479	4.79
^{210}Pb (Bq)	nm	nm	114	114	2.85
^{226}Ra (Bq)	19	75	7	101	0.51

nm = not measurable in tissue of either specimen

These estimated values are extremes assuming high consumption levels and no loss of radionuclides during cooking. Detection limit values, probably overestimating concentrations, are used in some cases. Furthermore, data for each species came from only two specimens and were selected because their tissue burdens for ^{226}Ra were high relative to others.

To indicate the magnitude of radionuclide intakes the values are compared to one tenth of the values of Annual Limits on Intake for occupational workers given by the ICRP (1979a,b). These values, which may be considered as maximum yearly radionuclide intake, are as follows: ^{210}Pb - 4000 Bq, ^{210}Po - 10,000 Bq, ^{226}Ra - 20,000 Bq, ^{228}Th - 50,000 Bq, ^{230}Th - 40,000 Bq, ^{232}Th 7,000 Bq (1,550 mg), ^{238}U - 80,000 Bq (6,500 mg). These levels approximate the Canadian regulatory dose limits for the members of the public. Clearly, estimated values of annual radionuclide intakes from consumption of grouse are considerably less than the limits on yearly intake.

As indicated, these are extreme estimates based on 'worst-case' assumptions concerning consumption rates and use values from only two animals, from the most contaminated areas, selected for their high levels of ^{226}Ra. It is considered unlikely that consumers would take in their diet even one tenth of the amount used in these calculations: grouse, although much appreciated by residents and sought after as a consequence, as mentioned previously, are difficult to obtain in numbers and are subject to legal season and catch (bag) limitations. Members of the Serpent River Band, located in the Low-Serpent area, probably consume more game, in relation to meat of domestic origin, in their diet, compared to that in the diet of Elliot Lake residents. However, this wild meat is likely to be substantially lower in radionuclides than that consumed by Elliot Lake inhabitants (if radionuclides occur in the same proportion to ^{226}Ra); grouse tissues there contain ^{226}Ra levels not significantly higher than in samples taken from undisturbed control areas. Assuming the more realistic consumption level of 7.1 $kg.y^{-1}$ then aggregate annual intake from wild game would be only 0.9% of the limit on annual intake in the case of grouse.

Quality Assurance - Samples of cow shank bone spiked with ^{226}Ra and ^{133}Ba, were run through the analytical procedure and showed recovery rates of 98 ± 10%. Standard samples from CANMET, similarly processed, gave results within 10% of the known values.

Duplicate estimates were performed blind on bone samples from two grouse from the experimental area at CANMET and also at the MONENCO laboratory. Results of the duplicate estimations are seen in Table 6. The difference in GEL 1 bone values cannot be accounted for.

CONCLUSIONS

The findings indicate that radionuclide contamination of grouse is very localized in the Serpent River drainage basin. Although grouse from Elliot Lake have bone radionuclide levels higher than distant controls (but similar to local controls), levels in muscle, liver, and kidney tissues showed no significant elevation associated with site. Levels of ^{226}Ra in diet and tissues of grouse are similar to those reported in other studies of animals and plants in the study area. Concentration ratios of ^{226}Ra between diet items and grouse tissues are similar to those reported for other animals in the area. As anticipated, levels of ^{226}Ra in gut contents reflect, in a general way, levels in diet items consumed in the study areas but the relationship is not precise enough to allow levels in gut contents to be used as a measure of environmental contamination.

Radionuclides other than ^{226}Ra are present in grouse tissues at low levels. Radionuclide intake by humans consuming even substantial amounts of grouse taken in the Serpent River drainage basin, are calculated to be less than annual limits set by the ICRP.

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Table 1 - Ra-226 in woody plants from study and control areas.

	[Ra-226] mBq.g^{-1} dry weight of tissue collection sites							
	Elliot Lake		Mid-Serpent		Low-Serpent		Control-dist.	
species	Leaves w(u)	Stems w(u)	Leaves w(u)	Stems w(u)	Leaves	Stems	Leaves	Stems
Trembling aspen *Populus tremuloides*	41.8(38.9)	68.9(55.6)	14.2(12.4)	11.0(9.0)	13.9	8.3	14.9	3.4
Largetooth aspen *Populus grandidentata*	52.7	98.7	-	-	3.7	4.7	-	-

^{a} sample small (<0.5g d.w.)
w washed, (u) unwashed
[ ] sample not partitioned
\- samples not taken or processed

Table 2 - Ra-226 in fungi from study and control areas.

	[Ra-226] mBq.g^{-1} dry weight of fungus					
collection sites:	Elliot Lake		Mid-Serpent		Low-Serpent	
species	w	u	w	u	w	u
Amanita	-	-	1.0	4.4	-	-
Fomes	8.9	11.8	-	-	51.7	48.7
Hydnum	2.7	-	-	-	-	-
Polyporus	215.4	-	8.2	10.4	2.6	1.3
Stereum	71.1	78.8	-	-	-	-
Trametes	-	-	-	-	2.1	2.7
Trichaptum	-	-	-	-	6.4	6.7

w washed,
u unwashed
\- samples not taken or processed

Table 3 - Levels of Ra-226 in water samples from study and control areas.

	[Ra-226] $mBq.l^{-1}$ water[a]				
collection sites:	Elliot Lake	Mid-Serpent	Low-Serpent Hwy 17 bridge	Low-Serpent Drive-In theatre	Control-distant
dissolved	118.1	122.4	95.1	80.1	12.1
total	558.7	192.0	232.0	358.7	9.0

[a] Average of two samples, each with duplicate subsamples

Table 4 - Radionuclides in tissues of grouse from Elliot Lake

	[radionuclide] in tissues,(dry weight basis)								
	AECL		MONENCO						CANMET
	^{238}U	^{232}Th	^{232}Th	^{230}Th	^{228}Th	^{210}Po[a]	^{210}Pb	^{226}Ra	^{226}Ra
	$\mu g.g^{-1}$		$mBq.g^{-1}$						
specimen tissue									
GEL 1									
bone	0.4	<1.0	<5.0	<5.0	<5.0	21.0±7.0	<50.0	10.0±5.0	150.2
muscle	0.2	<1.0	<5.0	<5.0	8.0±5.0	<5.0	<50.0	-	1.7
liver	0.2	<1.0	<5.0	<5.0	<5.0	15.0±5.0	<50.0	-	16.
GEL 5									
bone	<0.1	<1.0	<5.0	<5.0	<5.0	24.0±5.0	<50.0	<5.0	20.8
muscle	<0.1	<1.0	<5.0	<5.0	<5.0	7.0±5.0	<50.0	-	6.0
liver	1.0	<1.0	<5.0	<5.0	<5.0	16.0±5.0	50.0±30.0	-	3.9

[a] measured 4th May 1988

\- samples not taken or processed

Table 5 - Concentration ratios to tissue compartments of grouse from Elliot Lake.

COMPARTMENT	[Ra] ($mBq.g^{-1}$) dw	moisture %[b]	X_e = [Ra] ($mBq.g^{-1}$) ww	bone	muscle	liver	kidney
			COMPARTMENT:	bone	muscle	liver	kidney
			^{226}Ra concentration ($mBq.g^{-1}$)[a]				
			X_l =	17.3	1.0	3.2	7.8
				$f_{le} = X_l/X_e$ (concentration ratios)			
trembling aspen leaf	41.8	70.0	12.5	1.38	0.08	0.25	0.63
largetooth aspen leaf	52.7	70.0	15.8	1.09	0.07	0.20	0.50
fungi			24.5[c]	0.7	0.04	0.13	0.32
crop contents			8.2	2.11	0.13	0.39	0.96
chyme			9.1	1.90	0.11	0.35	0.86
chyle			5.6	3.09	0.18	0.57	1.40
water			0.12[d]	144.08	8.56	26.38	65.40

a wet-weight basis, calculated on log-transformed values, back-transformed mean values presented
b see text
c air-dry basis (see text)
d $mBq.g^{-1} \approx mBq.ml^{-1}$

Table 6 - Levels of Ra-226 ($mBq.g^{-1}$ dry weight)in grouse bone: repeated measures.

	[Ra-226] $mBq.g^{-1}$ dry weight of tissue	
sample	first measure	second measure
GEL 1 (grouse, Elliot Lake)	10.0 ± 5.0^{m}	142.5^{c}
GEL 5 (grouse, Elliot Lake)	$<5.0^{m}$	20.8^{c}

[c] measured at CANMET laboratory, Elliot Lake.
[m] measured at MONENCO laboratory, Calgary.

Table 7 - Average total ^{226}Ra levels ($mBq.L^{-1}$) (1984-1987) at water sampling stations in the experimental area in Elliot Lake (Anonymous, 1987b). Sample site numbers, location, map coordinates, average ^{226}Ra levels and sample size are indicated.

MOE Site #	Site Description	Latitude	Longitude	Avg total ^{226}Ra ($mBq.l^{-1}$) 1984-87	n
#001	Old Hwy 17 E of Hwys 108@17	46 12 40.9	82 30 43.92	75.6	20
#002	At lake Depot	46 20 7.52	82 32 22.78	21.6	20
#003	At Pecors Lake	46 22 26.74	82 26 16.91	85.3	11
#004	At Pecors Lake	46 23 36.85	82 29 54.14	165	13
#006	Crotch Lake	46 25 4.8	82 35 19.79	65.6	15
#007	Buckles Creek at Hwy 108	46 22 25.61	82 35 50.27	163.9	20
#009	Sheriff Creek at Hwy 108	46 24 9.12	82 39 49.8	72.5	20
#010	Rochester Creek Near Inlet to Quirke lake	46 29 57.97	82 31 24.36	54.3	7
#011	Serpent River near inlet to Quirke Lake	46 30 39.11	82 36 32.87	149	21
#012	Creek near road to Stanrock townsite	46 28 17.81	82 33 4.73	285	4
#014	Serpent R. at Panel Mine side rd	46 30 11.54	82 38 28.89	158	19
#017	Stollery L. at Denison dam	46 29 8.68	82 38 6.36	978	18
#019	Dunlop L. outlet	46 28 51.78	82 38 55.1	29.7	19
#020	Serpent R. Trib., Moose L. outlet	46 27 44.66	82 30 59.54	36.3	8
#023	Pronto Effluent at Hwy 17	46 12 6.4	82 41 52.59	87	19
#026	Serpent R. Trib. Panel Mine Tre plant outlet	46 30 27.99	82 32 21.51	314	23
#027	Elliot Lake Municipal Pumphouse	46 23 22.09	82 39 53.05	25	19
#030	Dunlop L. in Bay A	46 29 4.37	82 39 21.27	8	2
#031	Quirke L. SW of Stanrock Mine	46 28 6.32	82 34 14.73	77	3
#032	Quirke L. NE of CanMet Mine	46 29 13.97	82 31 44.24	80	2
#033	Quirke L. SE corner	46 28 20.44	82 31 49.77	75	2
#034	Quirke L. E of Denison Dam	46 29 10.87	82 35 31.64	75	2
#035	Whiskey L. S end Near Rum Point	46 24 27.28	82 20 56.9	60	2
#036	McCabe L., center of Lake	46 25 22.23	82 33 50.11	290	1
#037	Camp L., at S end	46 14 6.0	82 26 29.49	63	3
#038	Serpent Harbour, near Hospital Point	46 11 55.43	82 40 32.93	60	1
#039	McCarthy L., at W end	46 19 45.02	82 29 5.71	30	1
#040	McCarthy L., at S end	46 18 29.74	82 26 55.11	60	1
#041	Hough L., centre of lake	46 24 32.22	82 29 32.24	110	2
#044	Westner L. at ski club rd.	46 22 59.8	82 37 33.09	141	11
#045	Williams L. Creek, at Denison Mine rd.	46 29 44.31	82 38 7.43	163	12
#046	Pronto Ditch, outlet below Pronto Treat. plant	46 12 15.39	82 42 41.86	75	4
#049	Serpent R., at Quirke L. outlet	46 29 14.25	82 29 20.01	132	20
#051	Quirke Mine Tailings	46 30 30.32	82 39 14.5	178	19
#054	May L., S end of lake	46 25 38.35	82 28 51.88	220	1
#055	May L., N end of lake	46 26 42.52	82 29 40.48	120	1
#056	Panel Creek at Quirke L.	46 30 11.16	82 33 7.95	135	4
#067	Esten L., central part of lake	46 21 4.28	82 41 50.51	7	1
#070	Orient L. outlet	46 27 30.74	82 31 10.88	70	14
#071	Panel Mine Tailings Effluent	46 31 8.36	82 32 30.86	230	13
#072	Gravel Pit outlet	46 31 7.57	82 41 5.92	13	16
#073	Evans L. outlet	46 29 37.89	82 39 55.13	18	19
#074	Esten L. outlet	46 20 39.4	82 36 55.01	50	19
Intermediate stations					
##051 & 011	Top portion of Serpent R. from Quirke tailings			162.7	40
##009 & 044	Section encompassing Horne Lake			96.8	33
##007 & 074	Section encompassingNordic Lake			108.6	39
##014 & 011	Upper portion of Serpent R. to drainage at Quirke L			153.4	40
##067 & 074	Section encompassing Esten Lake			48.3	20

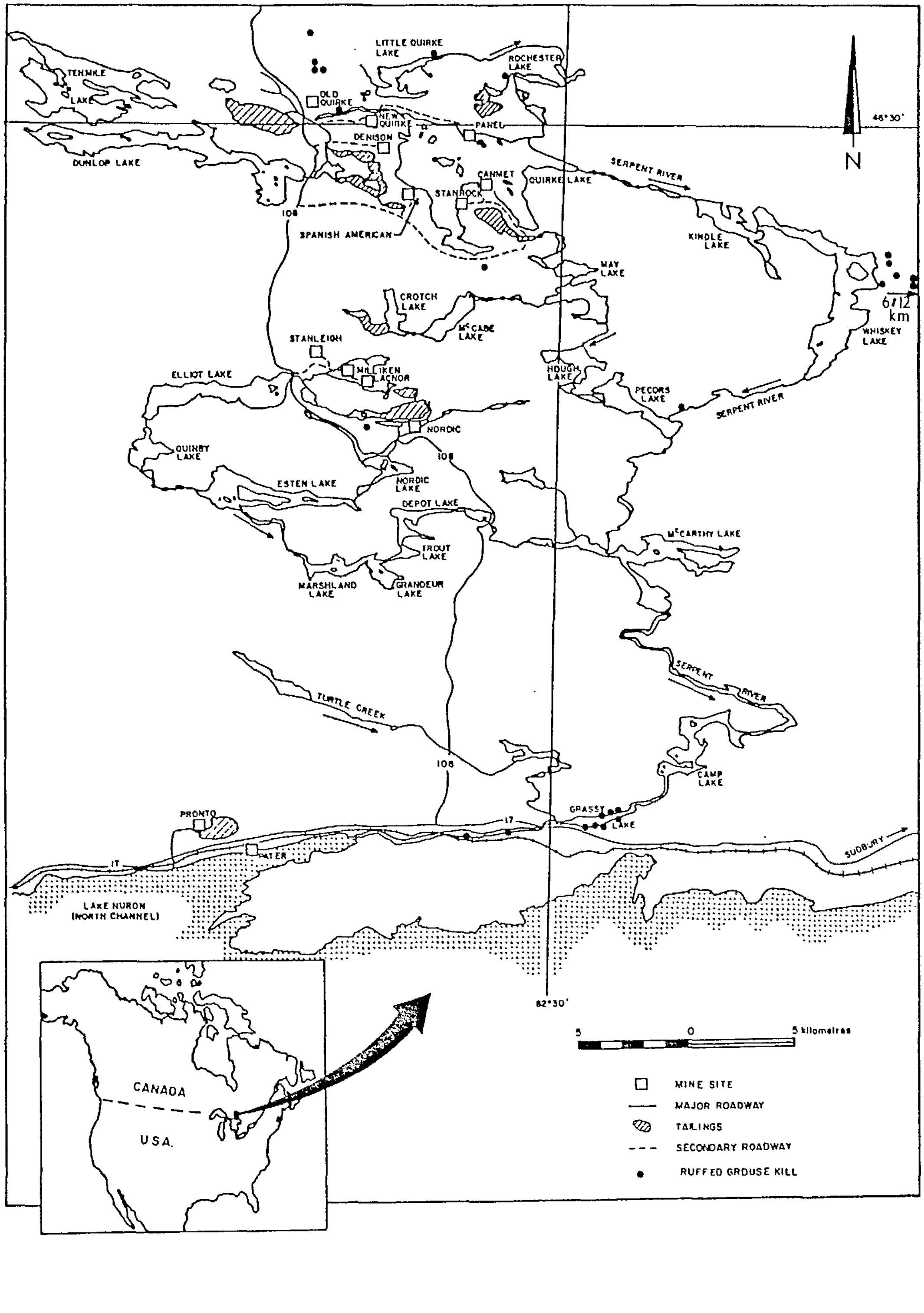

Fig. 1 - Location of grouse kill sites in the Serpent River watershed.

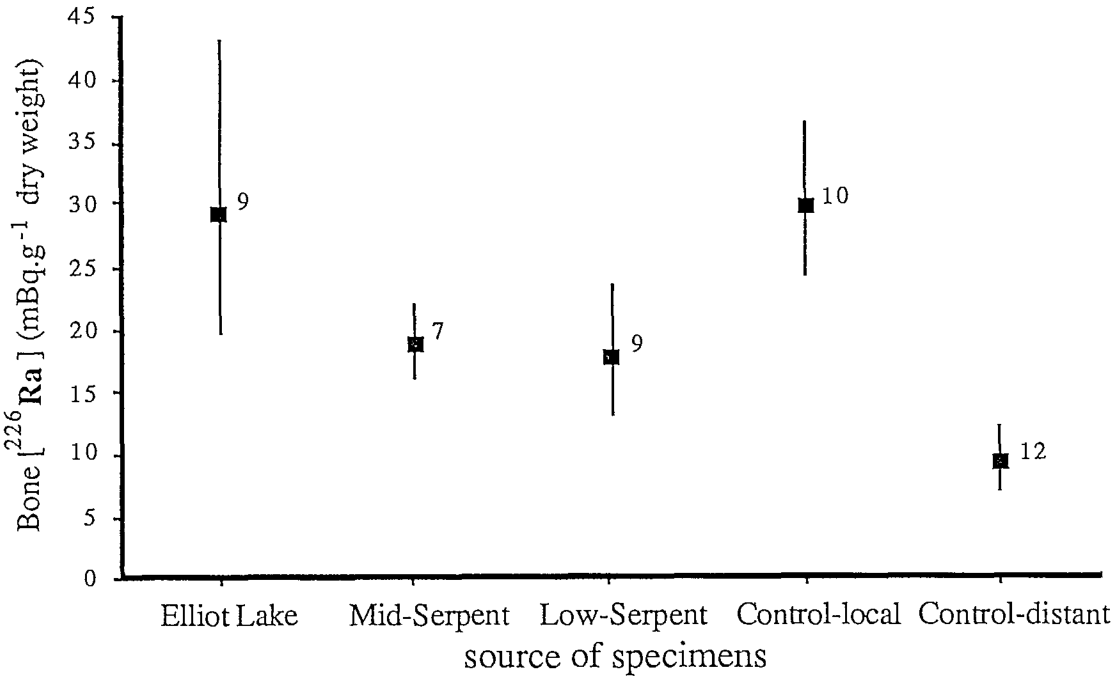

Fig. 2 - Levels of ^{226}Ra in bone tissue of grouse from study and control areas (mean ±1 SEM of log-transformed data calculated; back-transformed result, and sample size (n), shown in each case).

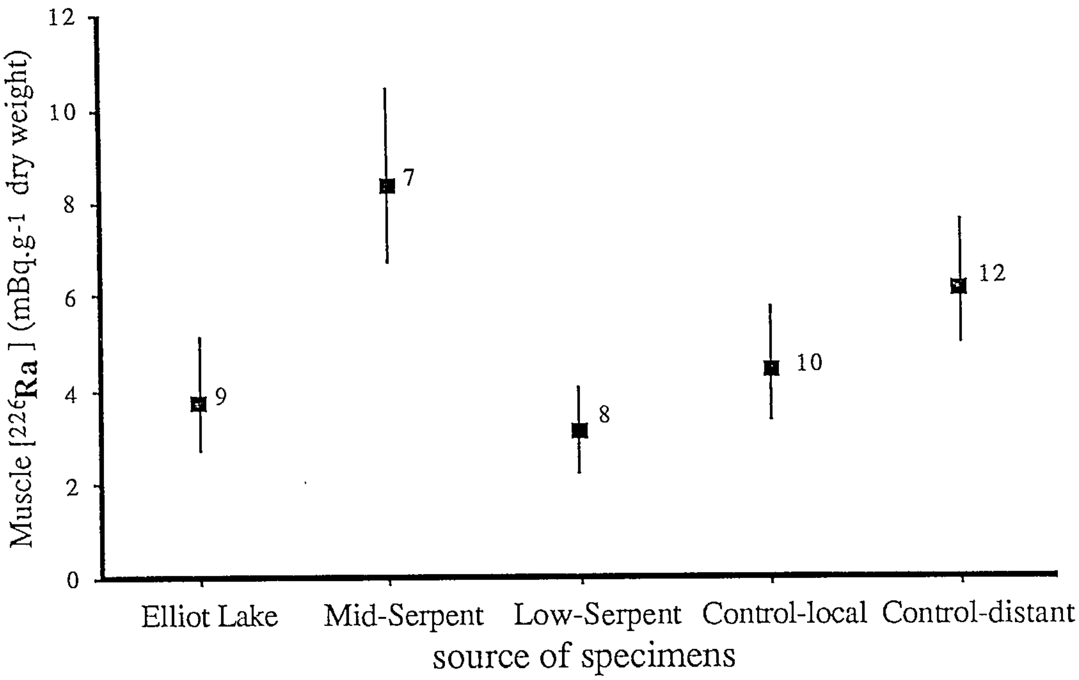

Fig. 3 - Levels of ^{226}Ra in muscle tissue of grouse from study and control areas (mean ±1 SEM of log-transformed data calculated; back-transformed result, and sample size (n), shown in each case).

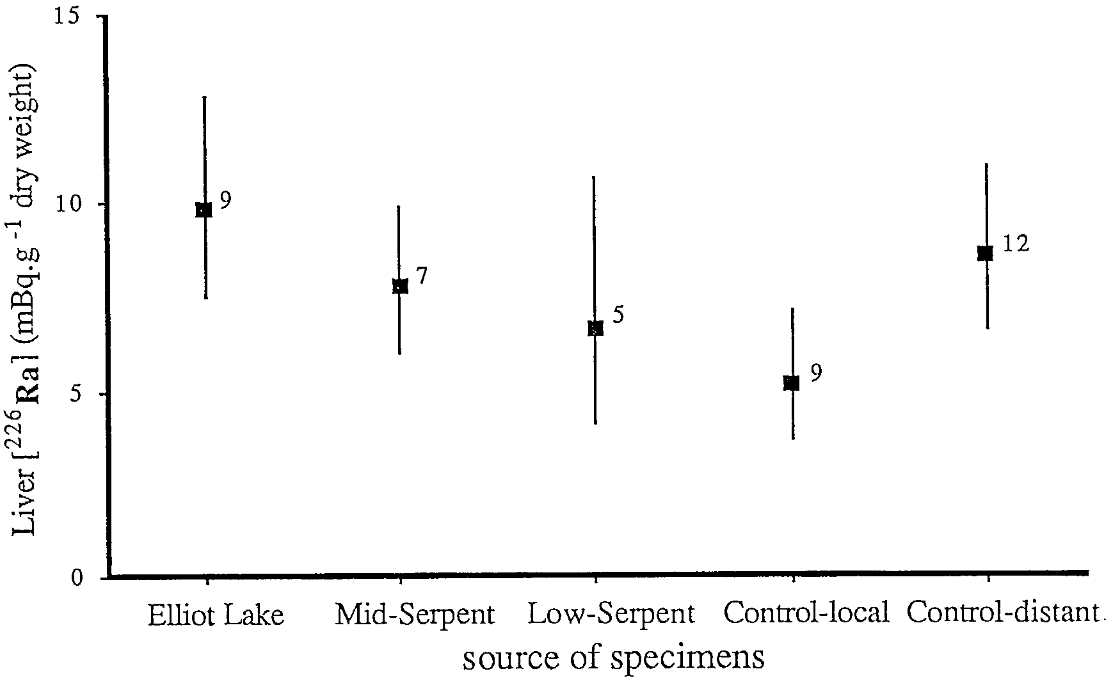

Fig. 4 - Level of ^{226}Ra in liver tissue of grouse from study and control areas (mean ±1 SEM of log-transformed data calculated; back-transformed result, and sample size (n), shown in each case).

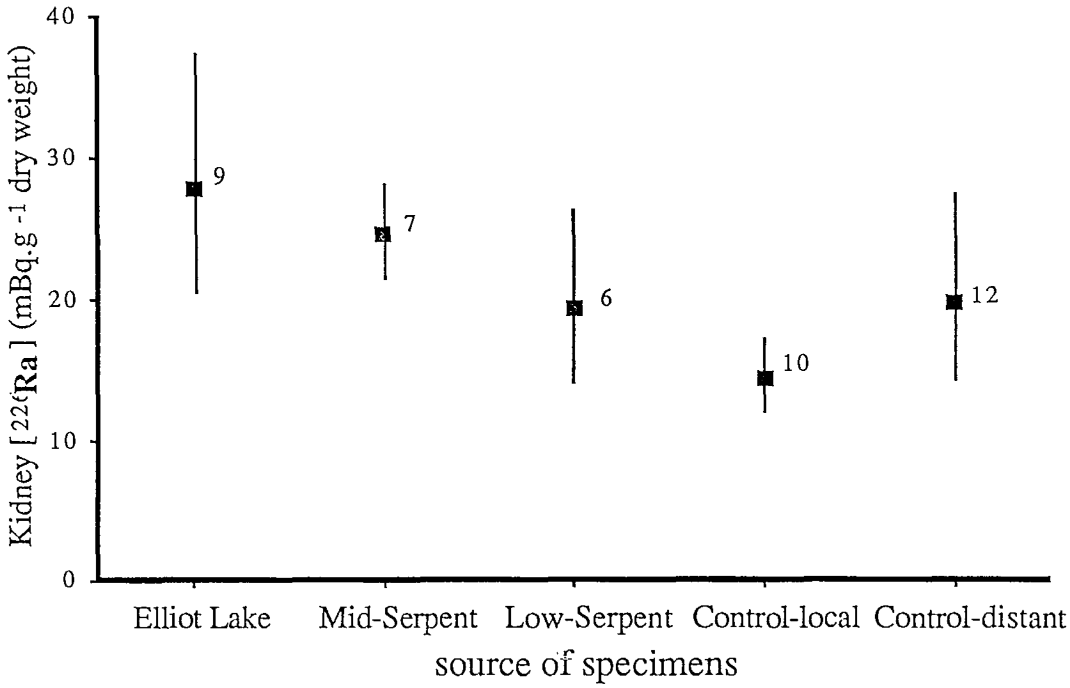

Fig. 5 - Levels of ^{226}Ra in kidney tissue of grouse from study and control areas (mean ±1 SEM of log-transformed data calculated; back-transformed result, and sample size (n), shown in each case).

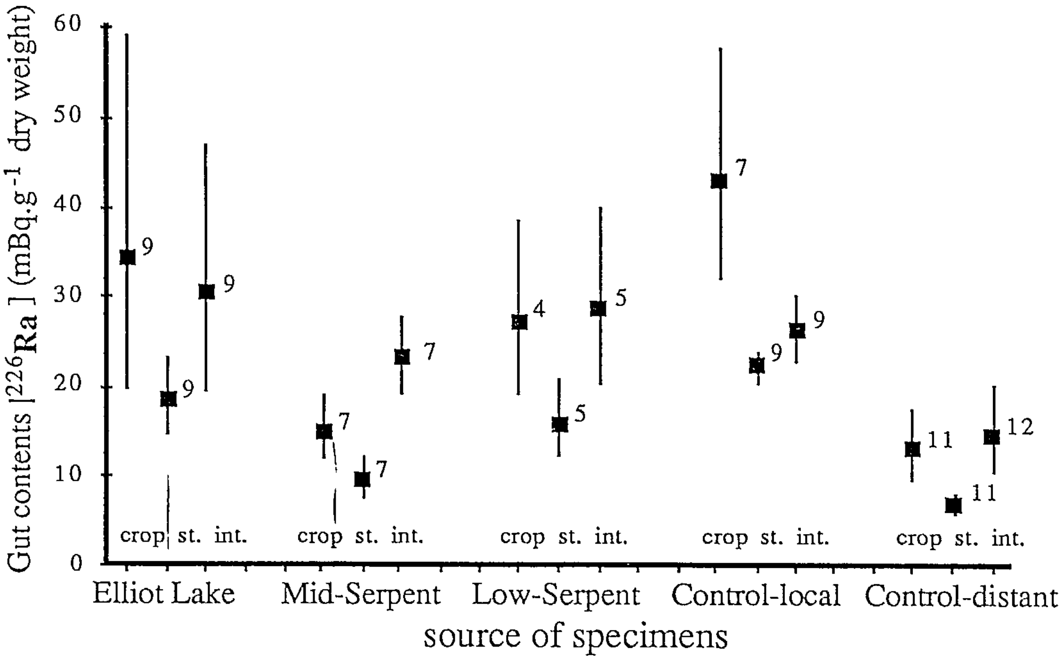

Fig. 6 - Levels of ^{226}Ra in gut contents of grouse from study and control areas (mean ±1 SEM of log-transformed data calculated; back-transformed result, and sample size (n), shown in each case).