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Ra-226 Concentration in Water Samples near Uranium Mines and in Marine Fishes

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RA-226 CONCENTRATION IN WATER SAMPLES NEAR URANIUM MINES AND IN MARINE FISHES

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บทคัดย่อ

การศึกษานี้เป็นการวิเคราะห์ปริมาณเรเดียม-226 และแคลเซียม ในตัวอย่างที่เก็บจากบริเวณใกล้เคียงเหมืองแร่ยูเรเนียม 3 แห่ง และในตัวอย่างปลาทะเล ที่เก็บจากอ่าว Puget Sound มลรัฐวอชิงตัน ประเทศสหรัฐอเมริกา ค่ามัธยฐานของปริมาณเรเดียม-226 ในตัวอย่างน้ำ เท่ากับ 0.428 พิโคคูรีต่อลิตร โดยมีค่าอยู่ในช่วงระหว่าง 0.043-1.552 พิโคคูรีต่อลิตร สำหรับแคลเซียมในตัวอย่างน้ำมีค่าระหว่าง 3.0 ถึง 190.0 มิลลิกรัมต่อลิตร สำหรับปริมาณเรเดียม-226 และแคลเซียม ในตัวอย่างปลาทะเล มีค่าตั้งแต่ 0.833-20.328 พิโคคูรีต่อกิโลกรัมน้ำหนักสด และ 114.1-259.3 มิลลิกรัมต่อกรัมเถ้า ตามลำดับ

ผลที่ได้จากการวิเคราะห์ พบว่า ปริมาณเรเดียม-226 ในตัวอย่างน้ำ มีค่าต่ำกว่าเกณฑ์กำหนดสูงสุดของกระทรวงสาธารณสุขและองค์การพิทักษ์สิ่งแวดล้อม แห่งสหรัฐอเมริกา ที่ยอมให้มีอยู่ในน้ำดื่ม นอกจากนี้ยังพบว่าปริมาณเรเดียม-226 และแคลเซียม มีความสัมพันธ์กันเฉพาะในตัวอย่างน้ำเท่านั้น ไม่พบว่ามีความสัมพันธ์อย่างเดียวกันในตัวอย่างปลาทะเล ยกเว้นในตัวอย่างปลา English sole

ABSTRACT

Radium-226 and Calcium were measured in water samples from the vicinity of three uranium mines and in fish samples collected from Puget Sound, Washington State. The radium content of the samples were below the maximum permissible concentration 3 pCi/L for drinking water recommended by the Public Health Service and U.S. Environmental Protection Agency. The mean value of Ra-226 in water was 0.428 pCi/L and ranged from 0.043 to 1.552 pCi/L, whereas calcium content ranged from 3.0 to 190.0 mg/L. Ra-226 concentrations and calcium content in whole fish were 0.833-20.328 pCi/kg wet wt. and 114.1-259.3 mg/g ash, respectively. Results of the study indicated that Ra-226 concentration in water was correlated with calcium concentration but that this correlation was not observed in fish sample except English sole.

I. Introduction

Radium-226 is a naurally occurring radionuclide and is extracted directly from uranium ore; therefore, it is released to ground and surface waters as natural leaching and as a water product from uranium mining. Ra-226 also occurs in rocks and soil as well as in the bottom sediments. Many benthic organisms such as worms and crustaceans ingest sediments and hence this radionuclide along with their food and link in the food chain. Fish according to the species may represent the second, third, or fourth trophic levels of the food chain. The Ra-226 content of fish species which are eaten by man will be more important. Since Ra-226 is retained in the bone, when taken into the body of man it damages the radiosensitive hematopoietic tissue in the bone marrow and may produce cancer. It is considered to be a very toxic radionuclide (Cember 1969).

The purpose of this study was to determine the concentration of Ra-226 in the ground and surface water collected in the vicinity of uranium mines and to examine the Ra-226 levels in marine fishes collected from Puget Sound in Washington State.

Calcium in the environment profoundly affects the physical and biological behavior of radium. Moreover, in the living systems, calcium isreferredtoas the "nutrient analog" of radium (Rope and Whicker 1985).Therefore,thisstudyincluded the analysis of calcium content.

II. Literature Review

A. Occurence

Radium is the heaviest member of the alkaline earth group. There are four naturally occuring isotopes of this element. The most important of these is the longest lived, radium-226, which is a member of the uranium-238 series as shown in figure 1. As a result of its relatively long half-life and the high natural abundance of its parent, Ra-226 is the most abundant radium isotope. Some of the physical properties reported for radium are summerized in table 1.

Radium-226 is present in uranium minerals and is a waste product of the uranium milling process. Mining of uranium can enhance radium release into water supplies by disturbing the uranium ore and exposing the ore surface to leaching processes. Because of its high solubility, Ra-226 can be found in water supplies far in excess of the concentration of the parent uranium (Sedet 1966).

B. Radium-226 in the environment

Radium-226 is widely distributed and is present in trace amounts in most materials. The range of Ra-226 concentration in vegetables in Italy and in Germany are 0.72-1.2 pCi/kg wet wt. (de Bortoli and Gaglione 1972) and 1.0-6.1 pCi/kg wet wt. (Muth 1960), respectively. The Ra-226 levels reported for the average diet in New York and San Francisco are 1.7-0.8 pCi/g Ca, with exclusion of drinking water (Fisenne and Keller 1970). Radium-226 content of worldwide soil sample is 0.08-3.8 pCi/g, excluding the high background areas, with a mean value of 0.72 pCi/g (de Bortoli and Gaglione 1972). Anderson, et al. (1963), reported that the natural background Ra-226 content in sediments of the Colorado River drainage averages 1.6 pCi/g.

The Ra-226 content of surface water is usually very low. Kaufmann and Bliss (1977) noted that the Ra-226 content of surface water ranged from 0.01 to 0.10 pCi/L, while some ground water may contain as much as 100 pCi/L. Anderson, et al. (1963), reported that the natural dissolved radium concentrations in surface water from an unpolluted location in the Colorado River basin average 0.2 pCi/L, but the concentrations in the Dolores River rose as high as 88 pCi/L in 1956 as a result of direct discharge of uranium mill wastes into the river. The average radium content of U.S. drinking water is 0.042 pCi/L (Hursh 1953). The concentration of Ra-226 reported for some rivers and lakes in Europe are as follow (de Bortoli and Gaglione):

Germany	0.07-0.84 pCi/L
Netherland	0.04-0.15 pCi/L
Yugoslavia	0.30-1.10 pCi/L

According to the same report, Ra-226 content in thermal and mineral waters in Italy are as high as 48 pCi/L. Polikapov (1966) reported a mean valve for radium content in U.S. rivers of 0.07 pCi/L, while in a tapwater it raged from 0.0 to 5.8 pCi/L. This is compared with the data reported for tapwater in Germany which is 0.03-0.34 pCi/L (de Bortoli and Gaglione 1972).

Edgington, et al. (1970), reported that the Ra-226 concentration in sand and in marine algae collected from coastal waters of Puerto Rico ranged from 0.033 to 0.086 pCi/g and from 0.045 to 0.073 pCi/g, respectively. The range of Ra-226 levels in ocean water is 0.08-0.15 pCi/L for bottom samples and approximately 10^{-4} pCi/L for surface sample (Eisenbud 1973).

C. Radium-226 in Fish

Radium-226 content for fish reported in the literature are listed in table 2. Swanson (1983) noted that fish from a lake affected by an operating uranium mine and mill had Ra-226 levels one to two orders of magnitude above level in fish from an uncontaminated control lake. In the same study, he found that Ra-226 was higher in the skin and bone than in flesh. This was attributed to the high calcium content of skin and bone and the fact that Ra-226 can replace calcium, resulting in high concentrations.

Calcium in the environment profoundly affects the physical and biological behavior of radium. The range of calcium in fresh waters (rivers and lakes) was given by Vanderploeg, et al. (1975), as 1.9-114 mg Ca/L. Krimholz and Foster (1957) reproted a range of 2-200 mg Ca/L. The mean calcium content of the river water of North America is 21.0 mg/L (Wetzel 1975), whereas in sea water it varies from 398 to 424 mg/l depending on the salinity (Edgington, et al., 1970)

The calcium content of fish has been investigated in a number of studies. Templeton and Brown (1964) reported calcium values from 48 to 75 mg Ca/g bone (wet weight) and 0.042-0.29 mg Ca/g muscle (wet weight) for brown trout (Salmo trutta) tissues. Vanderploeg, et al. (1975), reported a range of 19.5-80.4 mg Ca/g wet bone and 0.089-0.93 mg Ca/g wet muscle. The calcium content of blood of Atlantic salmon (Salmo salar) was reported by Hoar (1957) to range from 138 to 174 mg Ca/L and to depend on the life cycle stage of the fish.

D. Toxicity

The metabolism of Ra-226 in animals and plants is similar to that of calcium; it accompanies calcium throughout the food chain, finally being bound in the bone tissue. About 70-90 % of the Ra-226 in the body is contained in bones, the remaining fraction being distributed approximately uniformly in soft tissues. In animal studies, it has been found that although Ra-226 initially is deposited on bone surfaces, especially in areas of rapid bone formation , continual intake and bone remodeling cause the Ra-226 to become uniformly distributed throughout the bone (Wrenn, et al., 1985).

In 1930, a mixture of zinc sulfide (fluorescent substance) and Ra-226 was painted on watches and clocks by women workers. The workers often tipped the brushes on their lips or tongues. Years later, among 1,285 radium-dial women workers, 63 bone sarcomas and 23 head carnicomas were observed (Mays and Roland 1985). The effect of Ra-226 on mutation rates, and therefore on overall quality of the gene pool, is not well known.

The maximum permissible concentration of Ra-226 in drinking water recommended by the Public Health Service is 3 pCi/L, and by the Environmental Protection Agency (EPA) is 5 pCi/L (Lee, et al., 1980).

III. Materials and Methods

A. Sample Collections

Water samples were collected from certain locations in areas which may have received contaminated runoff from three uranium mines in northeastern Washington State. The mines are Western Nuclear, Inc., Dawn Mining Co., and Joy Mining Co. The study sites are shown on Fig.2.

The fish samples collected for this study were all bottomfish that are commonly consumed by man. Fish samples analyzed in this study are as follows:

1. Dover sole (*Microstomus pacificus*)
2. English sole (*Parophrys vetulus*)
3. Rex sole (*Glyptocephalus zachirus*)
4. Slender sole (*Lyopsetta exilis*)
5. Hake (*Merluccius productus*)
6. Longnose skate (*Raja rhina*)

B. Sample Preparation

Water samples were acidified with concentrated nitric acid to prevent radium adsorption and to remove the adsorbed radium from the glass surface as well. Barium carrier and barium-133 tracer were added to 2 liters of a water sample which was then evaporated to a small volume.

Fish samples were weighed, measured, sexed, and identified by species. Since it has been found from examinations made by the Public Health Service that fish can be effectively radioassayed in their entirety (Sabo, et al., 1963), the whole fishes were analyzed. Samples were dried at 80°C and dry-ashed at 550°C. Ashed fish samples were dissolved in concentrated nitric acid. Treatment with hydrogen peroxide and hydrochloric acid was sometimes necessary to remove unoxidized material and to clarify the solution. Barium-133 and barium carrier were used as tracer and carrier, respectively. The chemical yield was measured from Ba-133, whereas barium carrier was used to co-precipitate the radium.

C. Radium and Calcium Analysis

Radium-226 in the water samples was determined by the radon emanation technique. The radium was co-precipitated with barium sulfate. The precipitate was converted from sulfate to carbonate and dissolved in nitric acid. Chemical yield was determined by counting the gamma activity of barium-133. The solution was de-emanated and aged for two to four weeks to allow radon-222 to grow-in. The solution was again de-emanated into an evacuated scintillation cell, and the alpha activity of radon-222 was counted.

In the case of fish samples, precipitation was done the same way as described above, except that after the precipitate was converted to carbonate, it was dissolved in nitric acid. Radium daughter products were extracted from this solution into thenoyltrifluoroacetone. Barium sulfate was re-precipitated, stored for three weeks, and counted for Ra-226 alpha activity.

Calcium was determined by atomic absorption spectrophotometry in diluted aliquot portions. All the results have been corrected for the blank.

Calculation

$$\text{Ra-226 activity} = \frac{A}{BxCxDxExF}$$

where A = net alpha count rate (cpm)

B = sample volume (liters) or sample weight

C = efficiency of de-emanation and counting

D = correction factor $1-e^{-t}$ for radon-222 ingrowth, where t is the time from the first de-emanation of radon to the end of the second de-emanation

E = recovery of barium-133

F = correction factor $1-e^{-t}$ for radon-222 daughter ingrowth (Rn-222 decay), where t is the time from the end of the second de-emanation of radon to the time of counting

D. Quality Assurance

To ensure the reliability of the radium analysis techniques and the results, three different quality control methodologies were used.

1. Calibration with Ra-226 standards obtained from National Bureau of Standards (NBS)
2. Cross-checked analysis with U.S. Environmental Protection Agency (EPA) Quality Assurance program
3. Replicate determinations on the same sample

Also, to establish the accuracy of the results, sample spiked with known amounts of Ra-226 were analyzed.

IV. Results and Discussion

A. Radium-226 in water

The activity of Ra-226 and the concentration of calcium in water samples as well as the Ra/Ca ratio are given in Table 3. Surface and ground water samples were collected during the period from May 1981 throughout October 1984. Ra-226 concentrations found in the water samples are rather low; the mean value is 0.428 pCi/L and the range is 0.043-1.552 pCi/L. These values are lower than those reported by Rope and Whicker (1985), which ranged 12 to 23 pCi/L in water from a surface pond near an open-pit uranium mine. Calcium contents of the water varied over a wider range, 3.0 to 190 mg Ca/L, than the Ra-226 value. The linear regression model (Neter, et al., 1985) was used to examine the relationship between concentration of Ra-226 and calcium content. The correlation between Ra-226 and calcium, equal to 0.648, was highly significant (R = correlation coefficient = 0.42 for $df = 33$), since the t-test at 95% confidence level indicated a p-value less than 0.0005. The graph plot of Ra-226 contents is shonw in Figure 3.

Over the period of time collected, there were no major changes in the Ra-226 level for surface water samples at either the Dawn Mining Co. (DMC) sites or the Western Nuclear Inc. (WNI) sites, nor for ground water samples at the DMC sites as shown in Figure 4 and 5, respectively.

B. Radium-226 in Fish

In this study, 22 samples were collected on 12 September 1986 and 16 January 1987, and whole fishes were analyzed for Ra-226 and calcium. The results are reported in Table 4. Concentrations of Ra-226 ranged from 0.833 to 20.328 pCi/kg wet wt. and calcium contents ranged from 114.1 to 259.3 mg Ca/g ash. The mean Ra-226 and calcium values were 4.062 pCi/kg wet wt. and 199.3 mg Ca/g ash, respectively.

A regression analysis (Neter, et al., 1985) was performed on the Ra-226 content versus calcium content of the fish samples in the same way as for the water samples. The correlation coefficients calculated for all of the species analyzed were not significantly greater than zero, showing that there is no correlation between Ra-226 levels and calcium contents. However, when the data were analyze by species, this was not true for English sole (Parophrys vetulus); the correlation of Ra-226 and calcium is 0.916 with a correlation coefficient of 0.839 which makes the correlation significant at the 0.05 probability level ($p < 0.0005$). There are clear differences in the concentration of Ra-226 with increasing calcium. The graph plot of Ra-226 levels in English sole and calcium is shown in Figure 6.

No significant and consistent differences between sexes in any species were observed. Comparison of the Ra-226 activity among all sampling locations showed that slender sole (Lyopsetta exilis) and English sole (Parophrys vetulus) from Pt. Gardner Station E had the highest value. The Ra-226 concentrations of samples from this site were higher than most previously reported values for edible fish, which are around 1-10 pCi/kg wet wt. Muth, et al. (1960),

reported values of 4.0 pCi/kg wet wt. in herring and 6.3 pCi/kg wet wt. in haddock for Ra-226. A range of Ra-226 from 1.4 to 3.2 pCi/kg wet wt. was reported for perch (Perca fluviatilis) by de Bortoli and Gaglione (1972). However, the range of value measured at the Pt. Gardner Station E (16-20 pCi/kg wet wt.) is similar to levels in fish reported in other literature. Values reported by Jenkin (1969) in Pacific salmon (Oncorhynchus keta and O. kisutch) were 2-50 pCi/kg wet wt. Ra-226 content in brook trout (Salvelinus fontinalis) and rainbow trout (Salmo gairdneri) from surface ponds near an open-pit uranium mine was 11-43 pCi/kg wet wt. (Rope and Whicker 1985). Swanson (1983) noted that in lakes affected by a uranium mine and mill in Saskatchewan, Canada, the Ra-226 level in white sucker (Catostomus commersoni), lake whitefish (Coregonus clupeaformi), and lake trout (Salvelinus namaycush) were 9.5, 1.8, and 0.8 pCi/g ash, assuming 7.0%, 3.0% and 2.2% ash content of these species, respectively. The Ra-226 activity in this study versus fish species is shown in Figure 7.

The quality control of both procedures was tested for reproducibility and reliability using a water sample with Ra-226 obtained from EPA's Quality Assurance Division in Las Vegas, and spiked water samples. The data was shown in Table 5. The chemical yield and counting efficiency were determined by using a Ba-133 tracer and a standard radium solution. The chemical yield for both methods was consistently more than 85%. The lower limit of detection (LLD), blank value plus three times standard deviation for 30 and 60 minutes counted for emanation method and precipitation method were 0.12 and 0.27 pCi, respectively.

V. Conclusion

Radium has been found in waste products of uranium mining, in water supplies, and in the sediment. Radium has the lowest maximum permissible water concentration of any of the radionuclides in the uranium-radium series (Table 6), which indicates a greater hazard to man from ingestion of Ra-226 than the other members of the chain.

In this study, Ra-226 concentrations were measured in ground water and surface water samples collected in the vicinity of three uranium mines and in marine fish collected from Puget Sound, in Washington State. The fish in this study were confined to bottomfish; their usual habitat is on soft bottom and they feed mainly on clams siphons, other small molluscs, marine worms, small crabs and shrimps, and brittle stars. The fish samples in this study are of fish consumed by man; the flesh of sole (order Pleuronectiformes) and hake (Merluccius productus) being popular in British Colombia, as well as the pectoral fins of longnose skate (Raja rhina) (Hart 1980).

Concentration of Ra-226 in all water samples were below the maximum permissible concentration for drinking water (MPC_w), recommended by the Public Health Service and EPA as 3.0 pCi/l and 5 pCi/L, respectively. It can be concluded that there has not been any significant release of Ra-226 to ground or surface waters during the period of this study. Statistical analysis of the data showed that there is a correlation between Ra-226 and calcium content of water samples. In fish samples, This correlation was observed in English sole. The highest content of Ra-226 was found at Pt. Gardner Station E. But this value was within the range of value report in the literature.

VI. Recommendation

1. Periodic sampling of the water and marine fish should be conducted to continue evaluating possible trends in Ra-226 content over time.

2. Sea water should be analyzed for Ra-226 content, as the differences in Ra-226 content in fish can be explained partly by the differences in Ra-226 concentration in water.

3. The parameters such as concentrations of other elements which may interfere with radionuclide uptake; seasonal and annual temperature ; and the chemical speciation of the radionuclide should be considered.

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Table 1 Physical properties of radium (Sedet 1956).

Density	ca. 6 g/cm^3
Melting point	ca. 700°C
	ca. 960°C
Boiling point	ca. 1140°C
Ionic radius, Ra(II)	1.52 A
	1.37 A
Diffusion coefficient Ra(II) at 18°C	0.667 cm^2/sec
Equivalent conductance	57.3 cm^2/ohm
Ionization potential:	
First electron	5.26 eV
Second electron	10.10 eV

Table 2 Literature values for Ra-226 concentrations in fish

Type of sample	concentration	Reference
A. Flesh		
Rainbow trout, Salmo gairdneri	4-11 pCi/kg wet	Parsont (1967)
Various freshwater and pelagic fish	2 pCi/kg wet	Holtzman (1969)
White sucker, Catostomus commersoni	13 pCi/kg wet	Whicker (1972)
Pacific salmon, Oncorhynchus keta and O. kisutch	2-50 pCi/kg wet	Jenkin (1969)
Haddock	5.3 pCi/kg wet	Muth et al. (1960)
Cod	4.0 pCi/kg wet	Muth et al. (1960)
Yellow perch, Perca flavescens	1-10 pCi/kg wet	Parsont (1967)
Largemouth bass, Micropterus salmoides	1.5 pCi/kg wet	Parsont (1967)

Table 2 Literature values for Ra-226 concentrations in fish

Type of sample	concentration	Reference
B. Bone		
White sucker,		
Catostomus commersoni	1.2 pCi/kg wet	Whicker (1972)
Various pelagic and freshwater		
fish	0.05 pCi/g ash	Holtzman (1969)
Rainbow trout, *Salmo gairdneri*	3-12 pCi/g ash	Parsont (1967)
Yellow perch, *perca flavescens*	0.7-9 pCi/g ash	Parsont (1967)
Largemouth bass,		
Micropterus salmoides	0.4 pCi/g ash	Parsont (1967)
C. Whole Fish		
Perch, *Perca fluviatilis*	1.4-3.2 pCi/kg wet	De Bortoli and Gaglione (1972)
Lake chub, *Couesius plumbeus*	11.2-56.8 pCi/g ash	Swanson (1983)
White sucker,		
Catostomus commersoni	9.7-59.3 pCi/g ash	Swanson (1983)
Spottail shiner,		
Notropis hudsonius	51.8-70.2 pCi/g ash	Swanson (1983)
Ninespine stickleback,		
Pungitius pungitius	27.9 pCi/g ash	Swanson (1983)
Cisco, *Coregonus artedii*	4.7 pCi/g ash	Swanson (1983)
Trout-perch,		
Percopsis omiscomaycus	56.3 pCi/g ash	Swanson (1983)

Table 3 Ra-226 content of water near uranium mine.

Sample location		Ra-226 pCi/L	Calcium mg/L	Ra/Ca pCi/g
A. Surface Water				
DMC	SW1	0.101±0.022	4.7	21.5
		0.057±0.019	9.2	6.2
		0.124±0.032	13.1	9.5
	SW3	0.302±0.052	11.8	25.6
		0.271±0.043	20.3	13.4
	SW2D	0.136±0.033	190.0	0.72
		0.092±0.017	189.0	0.49
WN1	MW2	0.398±0.064	12.1	32.9
		1.552±0.113	20.4	76.1
		0.184±0.026	22.0	8.4
		0.231±0.033	21.1	10.9
	MW3	0.665±0.057	12.5	53.2
	MW4	1.204±0.077	14.9	80.8
		1.213±0.079	17.4	69.7
		1.232±0.093	14.3	86.2
		1.125±0.064	19.4	58.0
	MW6	0.611±0.047	25.9	23.6
		0.376±0.042	26.4	14.2
	PH	0.187±0.030	71.3	2.6
	L1	0.149±0.035	4.1	36.5
	L3	0.177±0.029	8.1	21.7
		0.222±0.056	4.9	45.4
JMC	Station 2	0.504±0.043	3.0	169.7
		0.854±0.094	9.6	89.3
B. Ground Water				
DMC	GW1	0.043±0.014	144.0	29.9
	GW2	0.411±0.038	179.0	2.3
	GW3	0.190±0.044	17.1	11.1
		0.334±0.048	20.1	16.2
		0.314±0.072	154.0	2.0

Table 3 Ra-226 content of water near uranium mine.

Sample location	Ra-226 pCi/L	Calcium mg/L	Ra/Ca pCi/g
GW6	0.371±0.043	19.6	19.9
	0.227±0.037	22.9	9.9
	0.348±0.049	162.0	2.1
	0.184±0.026	26.3	7.0
WN1 GW1	0.176±0.026	84.5	2.1

Table 4 Radium-226 content of whole fish collected on 12 September 1986 and 16 January 1987.

Species	Sample location, Pt. Gardner	Ra-226 pCi/kg wet wt	Ca mg/g ash	Ra/Ca pCi/g	Sex
English sole	Control site 1	5.466±1.199	213.6	0.5	F
Slender sole		4.933±0.900	259.3	0.6	F
Slender sole	Control site 2	1.809±0.591	182.8	0.3	M
Longnose skate		0.909±0.341	131.6	0.3	-
Rex sole	Navy site rep 1	8.921±1.895	194.3	0.6	F
English sole		2.590±0.586	197.3	0.4	M
Slender sole		2.406±0.563	228.0	0.4	F
Slender sole	Navy site rep 2	4.277±0.832	237.4	0.6	M
Slender sole		3.600±0.659	159.9	1.0	F
English sole	Control site 1	1.375±0.516	197.3	0.2	M
Dover sole	(Pt. Gardner)	0.833±0.220	219.6	0.3	-
English sole	Station E	20.328±2.623	207.9	1.5	F
Slender sole		17.000±3.165	124.0	0.6	M
Slender sole		15.934±3.049	237.1	0.7	F

Table 4 Radium-226 content of whole fish collected on 12 September 1986 and 16 January 1987.

Species	Sample location, Pt. Gardner	Ra-226 pCi/kg wet wt	Ca mg/g ash	Ra/Ca pCi/g	Sex
Hake		3.026±0.809	124.0	0.8	F
Hake	Station 3	2.274±0.581	114.1	0.8	F
Dover sole	Control OT	3.963±0.632	231.6	0.9	M
English sole		1.091±0.364	203.3	0.3	M
Slender sole		1.117±0.366	164.5	0.4	F
English sole	Transect 7 10N	2.915±0.712	215.6	0.4	F
Slender sole	Transect 10OM	0.899±0.337	247.6	0.2	M

Table 5 Reproducibility and reliability test.

Number of test	Results (pCi/L)
Emanation Method	
Spiked sample	
1	18.61
2	18.90
3	18.68
4	18.48
5	18.57
Average value	18.65±0.16
Known value	18.75
U.S. EPA sample	
1	5.97
2	6.02
3	6.03
Average value	6.01±0.03
Known value	6.00±0.90

Table 5 Reproducibility and reliability test.

Number of test	Results (pCi/L)
Precipitation Method	
Spiked sample	
1	8.62
2	9.77
3	9.54
4	8.83
5	10.01
Average value	9.35±0.60
Known value	9.38
U.S. EPA #1	
1	6.30
2	6.23
3	6.22
Average value	6.25±0.04
Known value	6.30±0.95
U.S. EPA #2	
1	5.99
2	6.13
3	6.06
4	6.10
5	6.19
Average value	6.06±0.08
Known value	6.10±0.92

Table 6 Maximum permissible concentrations in water for radionuclides in the uranium-radium series.*

Isotope	MPC_w in pCi/L	Critical organ
Ra-226	3.3	Bone
Pb-210	33	Kidney
Po-210	233	Bone
Th-230	667	GI tract
Th-234	6,667	GI tract
U-234	10,000	GI tract
U-238	13,300	GI tract
Bi-210	13,300	GI tract

(No value are given for other members of the series)

*1/30 Th HB 69 value of continuous occupational exposure as recommended by the International Commission on Radiation Protection. The U.S. Public Health Service Drinking Water Standards (1962) limit the Ra-226 concentration in water used for drinking to a yearly 3.0 pCi/L

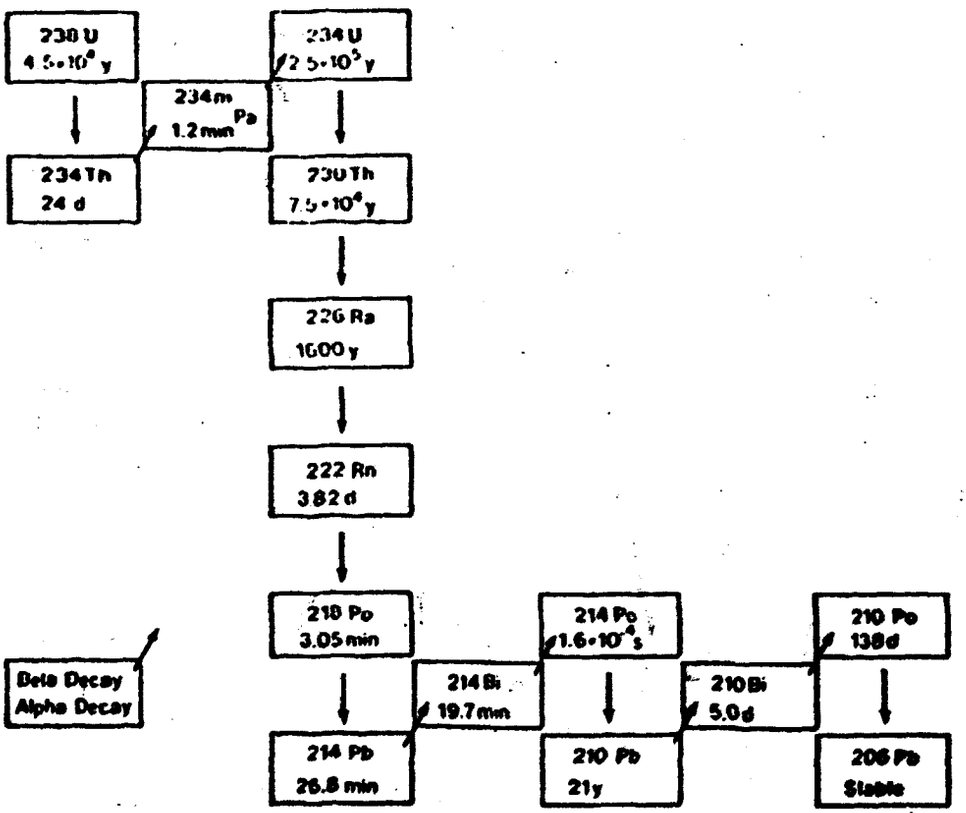

Fig. 1. Uranium series decay scheme.

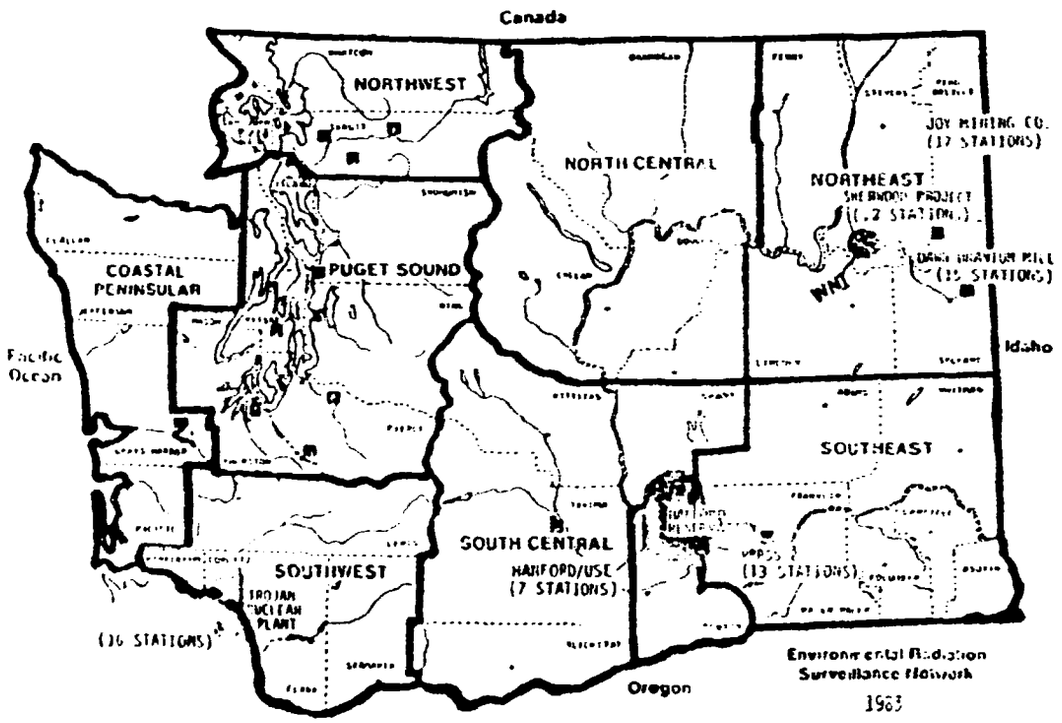

Fig. 2. Location map of study area for water and fish samples collected.

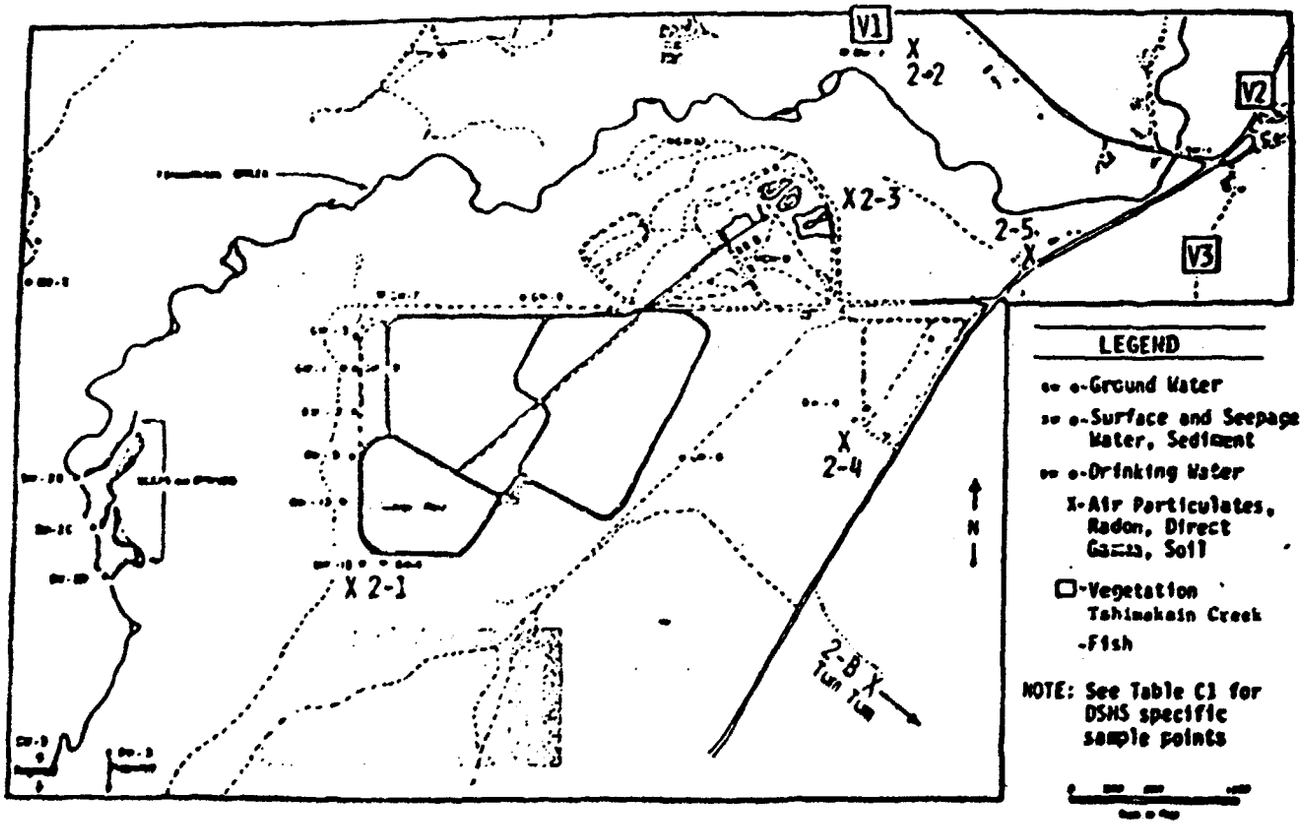

Fig 2.1 Sampling stations at Dawn Mining company (DMC)

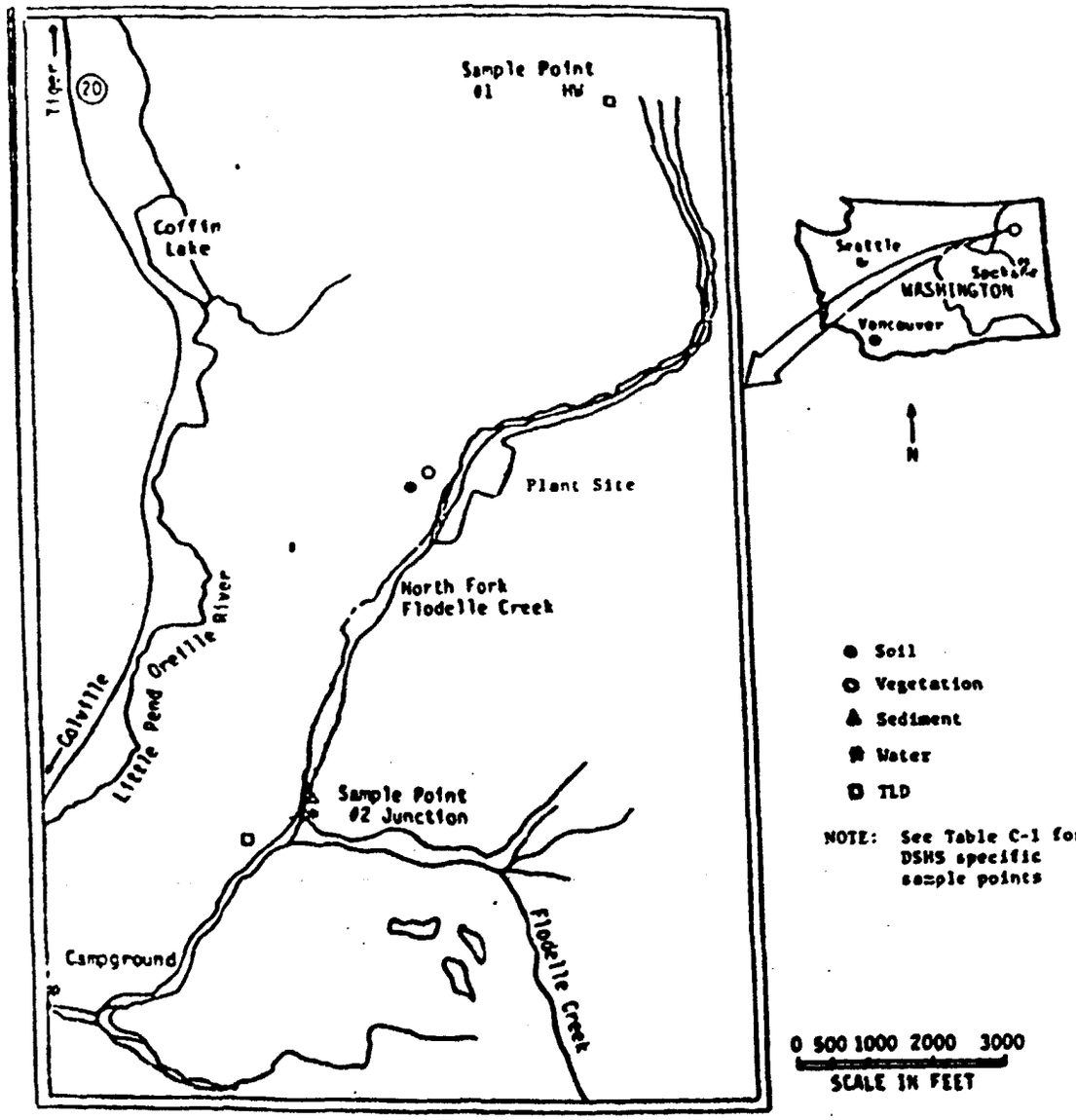

Fig 2.2 Sampling stations at Joy Mining company (JMC)

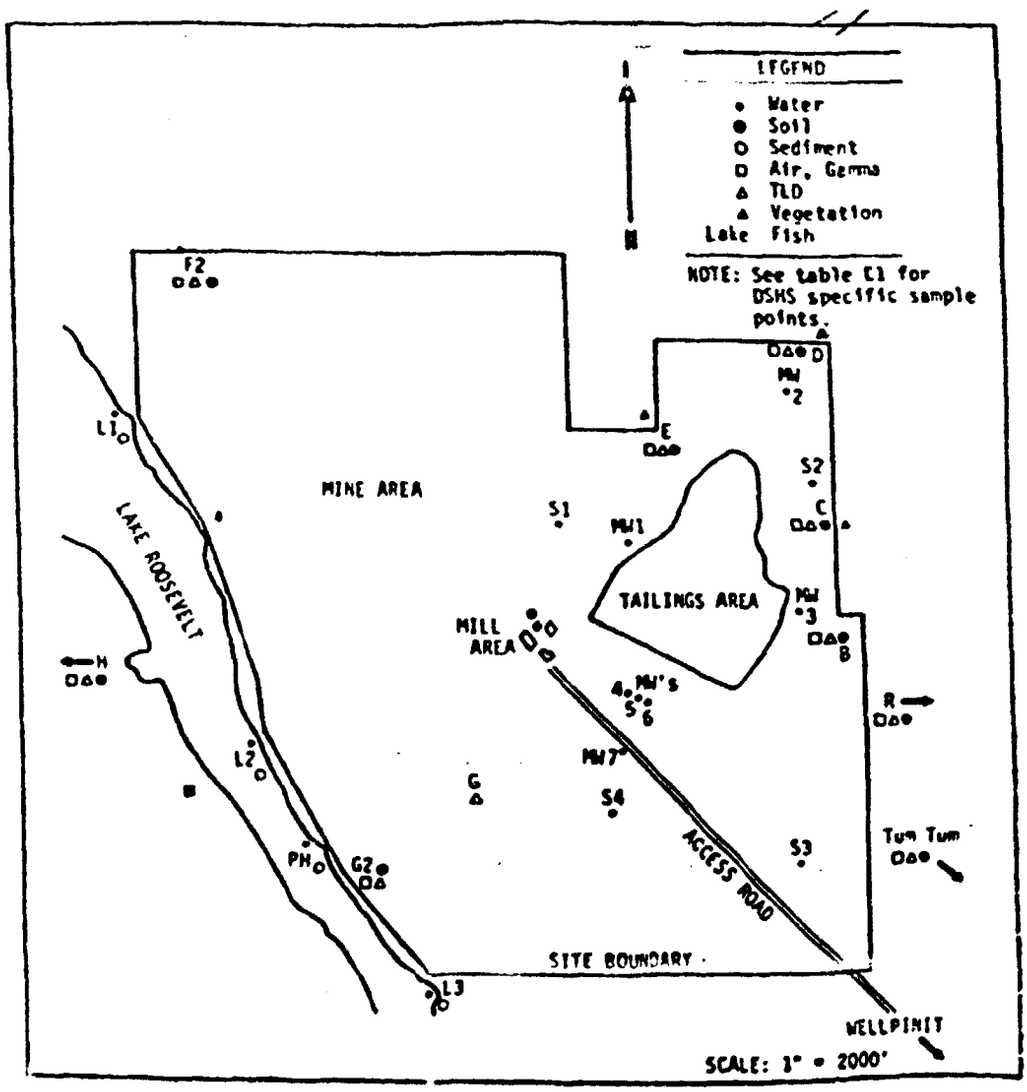

Fig 2.3 Sampling stations at Western nuclear, Inc. (WNI)

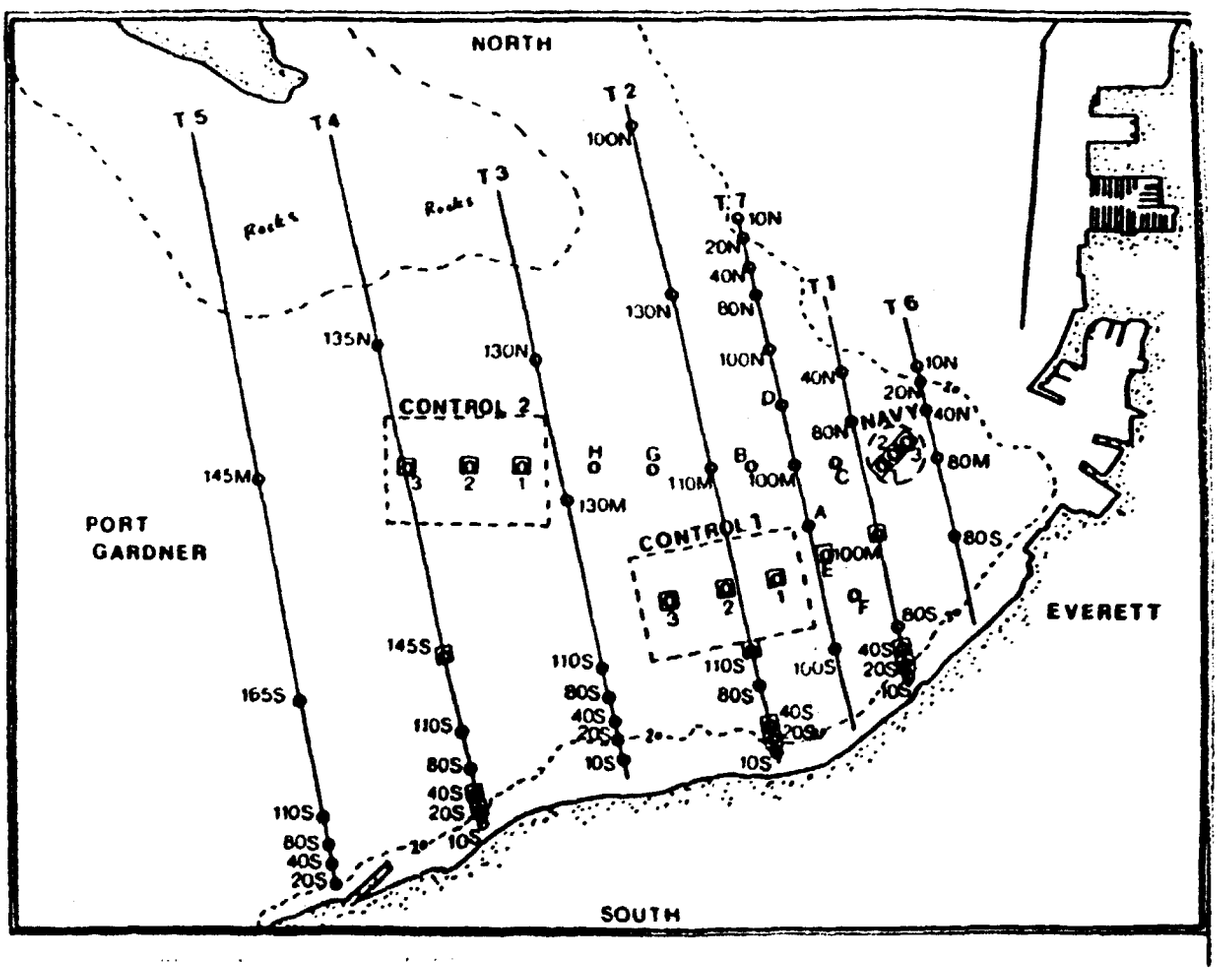

Fig 2.4 Sampling stations in Port Gardner
Depths in meters. N = North, M = Middle, S = South

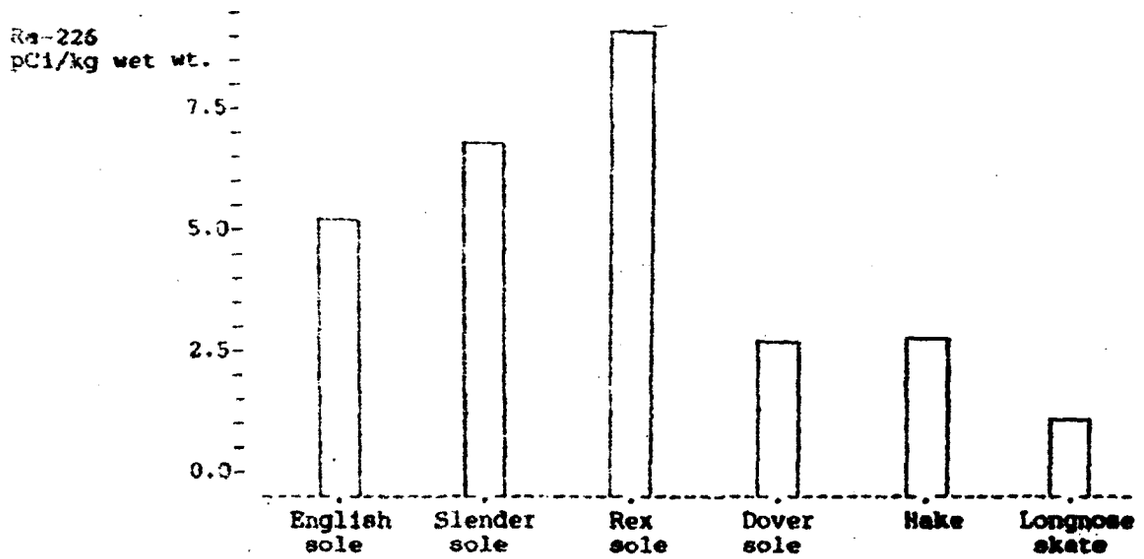

Fig 7. Ra-226 level (pCi/kg wet wt.) vs. species

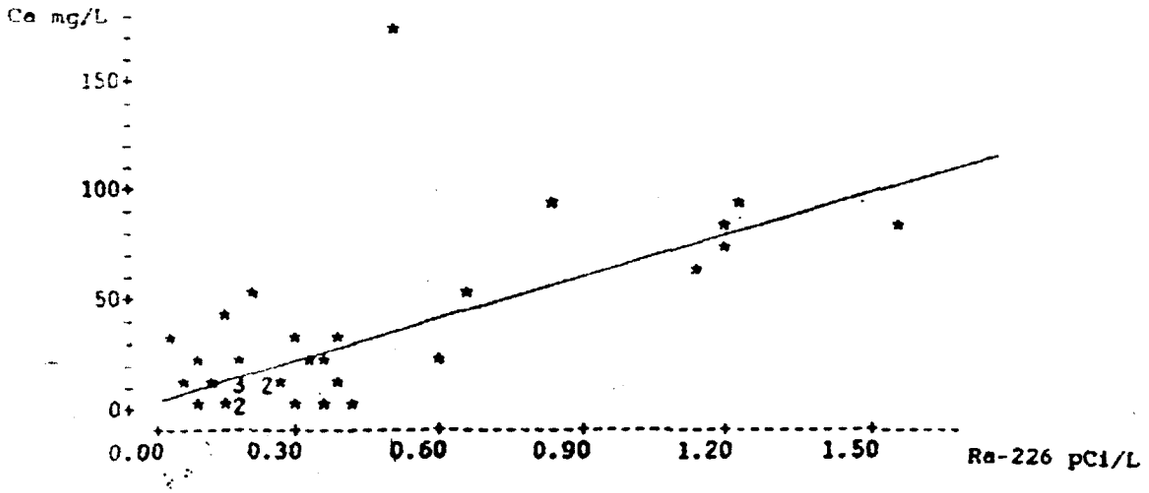

Fig. 3. Calcium contents of water samples vs. Ra-226 level.

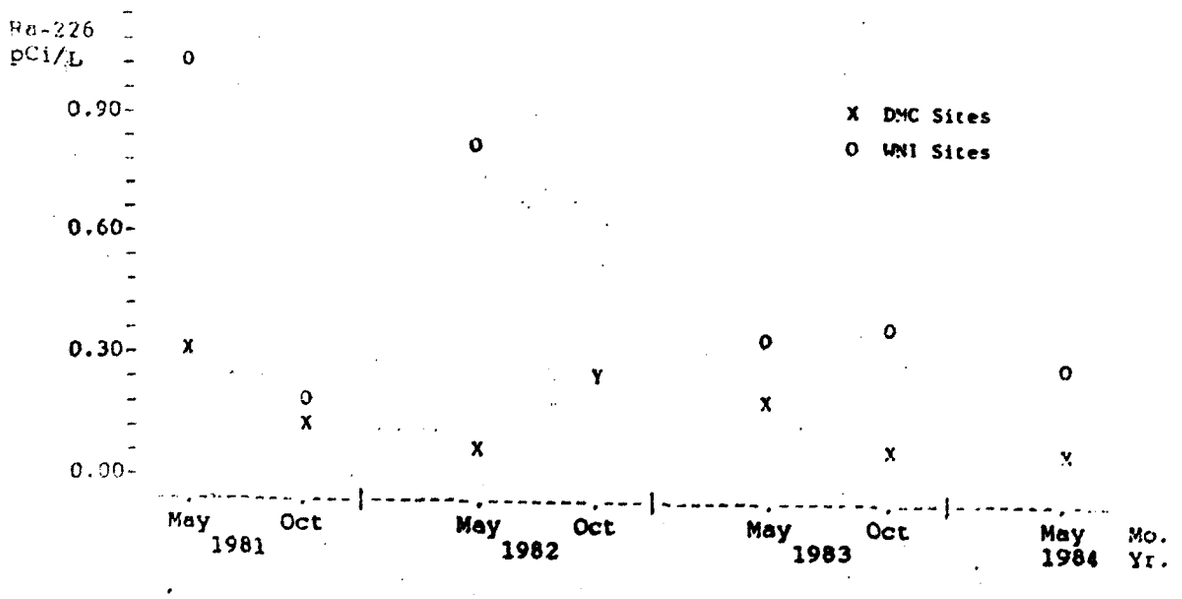

Fig 4. Ra-226 contents of surface water sample at DMC and WNI sites (pCi/L) vs. period of time collected.

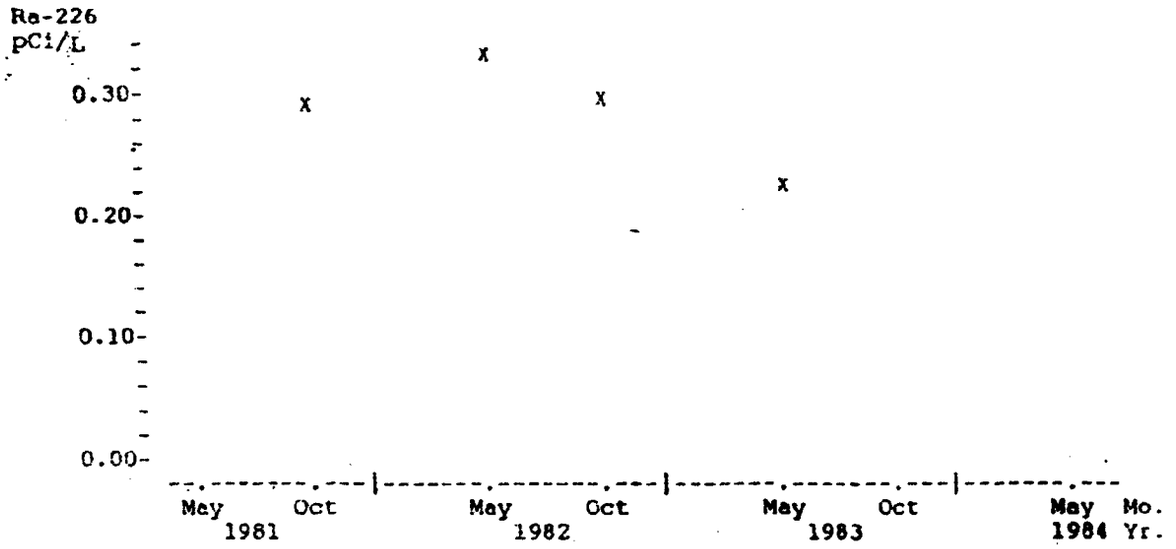

Fig 5. Ra-226 contents of ground water collected at DMC sites (pCi/L) vs. period of time collected.

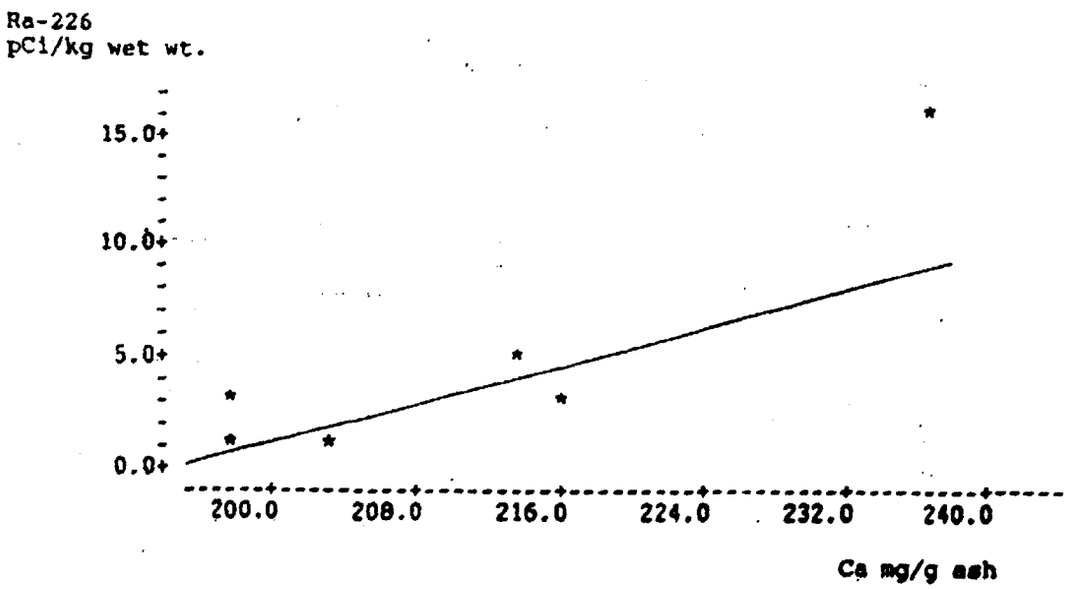

Fig. 6. Ra-226 contents of English sole (Parophrys vetulus) in pCi/kg wet wt. vs. calcium contents in mg/g ash.