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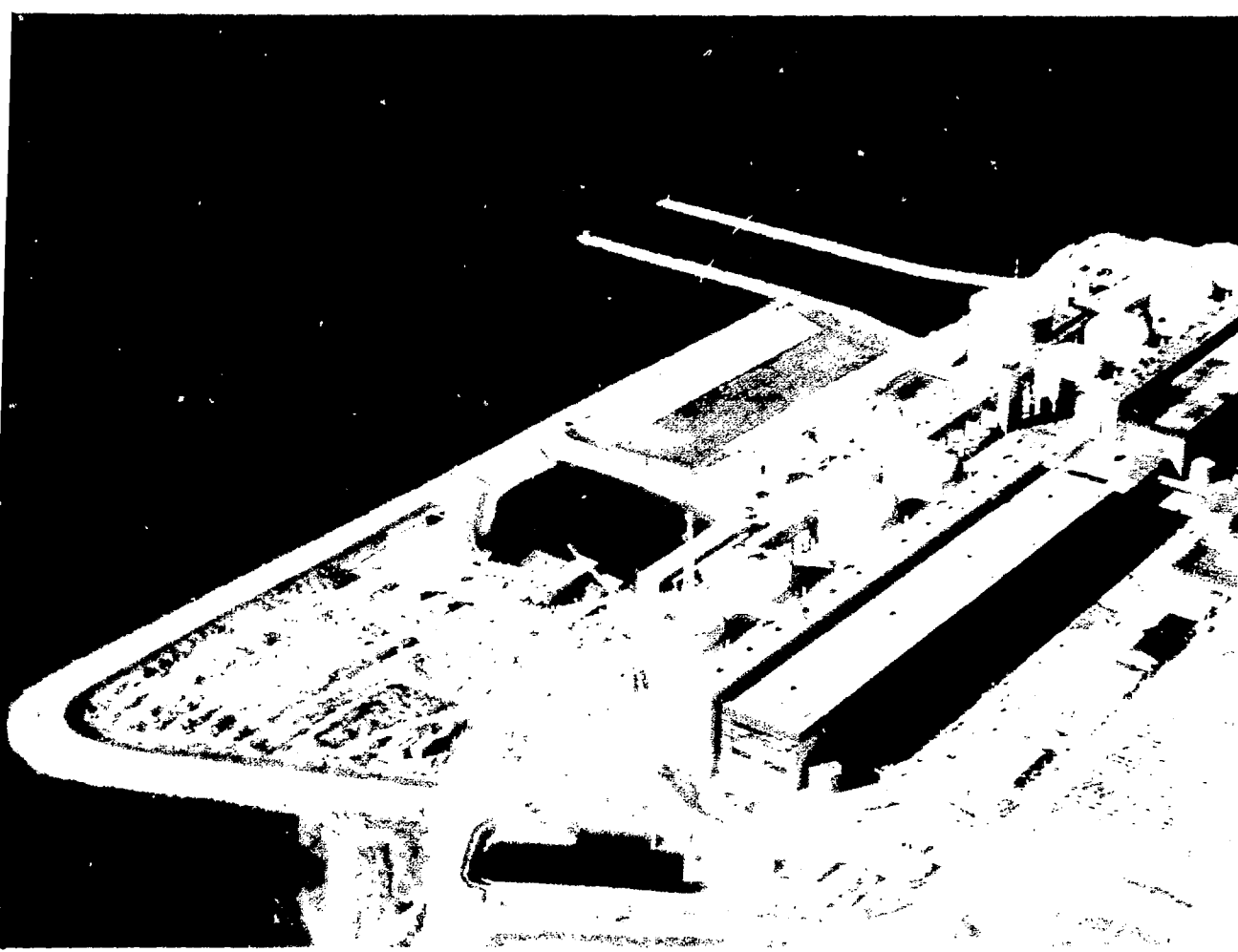


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Radionuclide Content of Canadian Surface Waters: A Report on the National Radionuclides Monitoring Program, 1981-1984

A.S. Baweja, S.R. Joshi and A. Demayo



SCIENTIFIC SERIES NO. 156

**INLAND WATERS/LANDS DIRECTORATE
WATER QUALITY BRANCH
OTTAWA, CANADA, 1987**

Canada

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Cover photograph–

Nuclear power station at Pickering, Ontario. (*Courtesy of Atomic Energy of Canada Limited*)



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Abstract

The National Radionuclides Monitoring Program was initiated to assess the radiological status of surface waters at a limited number of sites across Canada. During four years of operation (1981 to 1984), up to 13 locations were sampled monthly for water and two for sediments in 1982. Most water samples were composited annually, but some were composited quarterly to assess the temporal variations. The composites were then analyzed for total U, Ra-226, Cs-137, Sb-125 and tritium.

Levels of the radionuclides monitored are generally low, and no time trends were discernible during the monitoring period. Temporal variations among the quarterly analyses are mostly insignificant. Except for total U at certain locations, all other radionuclide levels were much below the "target concentration" guidelines for Canadian drinking water. The main contribution to the radiological dose commitment was from total U and Ra-226. This dose was below the objective dose of 10 μ Sv (1 mrem) as stipulated in the Great Lakes Water Quality Agreement of 1978. Contributions from individual radionuclides to cumulative dose were in the order: total U > Ra-226 > tritium > Cs-137 > Sb-125.

The limited sediment data suggest that the nuclear facilities along the Ottawa River, which are the nuclear power generating station (NPD) at Rolphton and the Nuclear Laboratories of the Atomic Energy of Canada Limited at Chalk River, have increased the levels of Cs-137 and Co-60 but have not affected those of Pb-210 and Ra-226.

Total U and Ra-226 activities data suggest disequilibrium; the levels of total U in water were much higher than those of Ra-226. Concentrations of the two radionuclides, however, were positively correlated for most surface waters.

Résumé

Le Programme national de contrôle des radionucléides a été mis en oeuvre pour évaluer la radioactivité des eaux de surface dans un nombre limité d'endroits un peu partout au Canada. Durant les quatre années de son application (de 1981 à 1984), on a prélevé mensuellement des échantillons d'eau à 13 endroits au plus et des échantillons de sédiments à deux endroits en 1982. La plupart des échantillons ont été combinés sur une base annuelle, mais certains l'ont été sur une base trimestrielle dans le but d'évaluer les variations avec le temps. On a dosé le U total, le Ra-226, le Cs-137, le Sb-125 et le tritium dans ces échantillons composites.

Les concentrations de radionucléides étaient généralement faibles, et on n'a discerné aucune variation avec le temps durant la période de contrôle trimestrielle. À l'exception du U total à certains endroits, les concentrations de tous les autres radionucléides étaient bien inférieures à la «concentration cible» stipulée dans les lignes directrices pour la qualité de l'eau potable au Canada. L'U total et le Ra-226 étaient les radionucléides qui contribuaient le plus à la dose engagée. Cette dose était inférieure à la dose visée de 10 μ Sv (1 mrem) stipulée dans l'Accord de 1978 sur la qualité de l'eau dans les Grands lacs. La contribution de chaque radionucléide à la dose cumulée diminuait dans l'ordre suivant : U total > Ra-226 > tritium > Cs-137 > Sb-125.

D'après des données limitées sur les sédiments, les installations nucléaires situées sur la rivière des Outaouais, c'est-à-dire la centrale nucléaire de Rolphton et les laboratoires nucléaires de l'Énergie atomique du Canada Limitée à Chalk River, sont responsables d'une augmentation des concentrations de Cs-137 et de Co-60; par contre, les concentrations de Pb-210 et de Ra-226 sont restées inchangées.

Les données sur la radioactivité de l'U total et du Ra-226 laissent supposer l'existence d'un déséquilibre; les concentrations de U total dans l'eau étaient de beaucoup supérieures à celles du Ra-226. Cependant, on a établi, pour la plupart des eaux de surface, une corrélation positive entre la concentration de ces deux radionucléides.

Radioactivity Units

The International System of Units (SI) has been used in this report. Basic SI radioactivity units are defined as follows.

becquerel (Bq): A unit of measure of radioactivity which corresponds to one disintegration (transformation) per second (dps).

gray (Gy): This unit is a measure of the biological effectiveness of radiation dose which represents the absorption of one joule of energy per kilogram of tissue.

sievert (Sv): The radiation dose equivalent is expressed in sieverts and is obtained by multiplying the absorbed dose with a quality factor (Q), which is variable depending upon the nature of radiation. For example, the value of Q is 1 for beta particles and gamma rays, whereas for alpha particles it has a value of 20 (U.S. EPA, 1981). N represents any other modifying factors, and is usually assigned a value of unity.

EQUIVALENT UNITS

1 curie (Ci) is equal to 3.7×10^{10} dps, or Bq

1 rad is equal to 100 ergs/g, or 0.01 Gy

1 rem is equal to 100 ergs/g \times Q \times N, or 0.01 Sv

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INTRODUCTION

Water Quality Branch Monitoring Mandate

Canada is blessed with abundant surface water supplies. In order that water of good quality be available for present and future uses by all Canadians, judicious management programs must be maintained. The Water Quality Branch (WQB) is responsible for providing scientific and technical information, and advice relating to water quality (Water Quality Branch, 1985a). Quality affects the suitability of water for human consumption, fish and other aquatic life, recreation, irrigation, livestock watering and industrial uses. A current high departmental priority is the management of toxic chemicals such that their detrimental effects on the Canadian environment are reduced, or eliminated. Radionuclides are some of the toxic chemicals of concern.

Sources of Radionuclides

Radionuclides are present in the aquatic environment as a result of natural sources and man's activities (Whicker and Schultz, 1982). The major global contribution of man-made radionuclides has largely come from fallout resulting from nuclear weapons testing in the atmosphere. Fallout inputs, however, have significantly decreased since the Limited Nuclear Test Ban Treaty of 1963, which banned large-scale atmospheric testing of nuclear weapons.

The weathering of naturally occurring radioactive minerals containing uranium and thorium series radionuclides (Figs. 1 and 2) usually contributes low concentrations of these radionuclides to large water bodies as can be seen from the data for Lakes Ontario (Durham and Joshi, 1980a) and Huron (Durham and Joshi, 1980b). Nevertheless, elevated concentrations of naturally occurring radionuclides can occur in fresh waters due to rich uranium deposits near the surface (B.C. Royal Commission, 1980) and/or nuclear fuel cycle operations (Hart and McKee, 1985; Platford *et al.*, 1984).

The interaction of cosmic rays with the atmospheric nuclides produces a variety of radionuclides; the overall proportion of such nuclides in the aquatic environment, however, is rather small. Radionuclides present in the atmosphere are generally brought down to the aquatic environment via precipitation scavenging and by dry fallout. Industrial and medical applications of commonly used radionuclides seem to contribute very little to the radiological water quality (Durham and Joshi, 1979a).

Table 1 lists the basic properties of some environmentally significant radionuclides released to the aquatic environment at various stages of nuclear activities. Many of these are also present in the fallout from nuclear weapons testing in the atmosphere.

Environmental and Health Impacts

There are over 200 radionuclides present in the environment. The ionizing radiation — alpha, beta and/or gamma — emitted by these nuclides varies in its physical properties and interacts with the biological matter differently; alpha particles are the most damaging because of their high energy. In this report, results of the measurements and calculated radiation dose are expressed in SI units.

Excessive ionizing radiation is viewed as potentially harmful to humans and biota alike. For example, intense radiation in excess of 10 sieverts (Sv) to the whole human body in a short period of time can be fatal (Johnson and Tutiah, 1985; U.S. EPA, 1981). Lower doses may not be immediately fatal but can cause latent adverse health effects such as initiation of cancer and/or genetic changes. Adverse health effects are generally proportional to the dose received by an individual. However, at low doses, there is a controversy in the scientific literature concerning whether or not there exists a threshold level (stochastic vs. non-stochastic effects) below which no ill health effects are discernible (ICRP, 1977, 1979; Johnson and Tutiah, 1985; U.S. EPA, 1981; Whicker and Schultz, 1982). Generally,

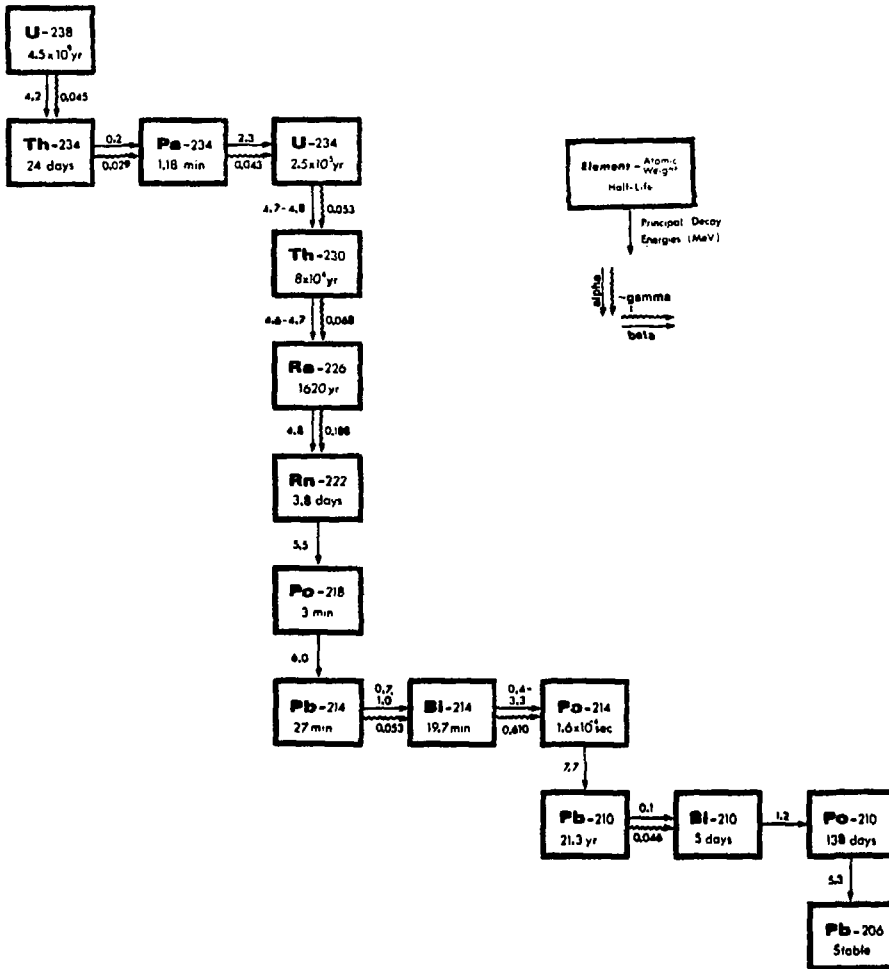


Figure 1. Uranium-238 decay chain series radionuclides with half-lives and mode of decay (Source: B.C. Royal Commission, 1980).

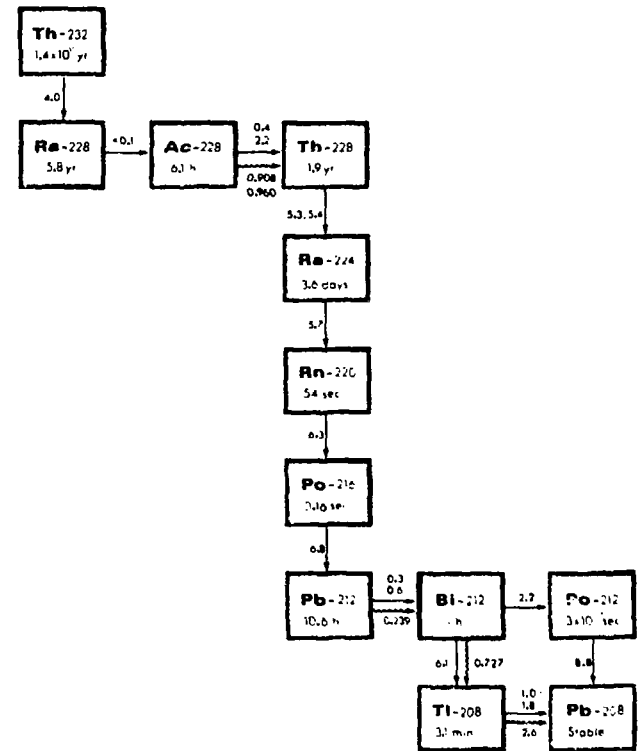


Figure 2. Thorium-232 decay chain series radionuclides with half-lives and mode of decay (Source: B.C. Royal Commission, 1980).

organizations responsible for radiation protection use the linear hypothesis for setting standards and guidelines.

The most significant pathway to man for the radionuclides contained in water is through its consumption as drinking water (U.S. EPA, 1981). Once ingested, the radionuclides usually migrate to certain organs of the body, the so-called "critical organs." For example, radium is deposited in the bone tissue, whereas uranium is lodged in the kidneys. The next important aquatic pathway involves consumption of fish which are known to accumulate radionuclides (Joshi, 1984).

In Canada, the Atomic Energy Control Board (AECB) is responsible for establishing dose limits. It is recommended by the Board that atomic radiation workers may not exceed an annual dose of 50 mSv, while the dose for a member of the public is set at 5 mSv per year. This dose is in addition to what is receivable as background radiation, which is about 2 mSv per year in Canada (Johnson and Tutiah, 1985). Since the effects of ionizing radiation on biota are cumulative, the AECB recommends that the radiation exposure should be kept as low as reasonably achievable -- the ALARA Principle.

Table 1. Basic Properties of Environmentally Important Radionuclides*

Radionuclide	Half-life†	Yield (%)	Decay mode	Comments
<i>Fission Products</i>				
Sr-90	28.5 y	5.9	β	Chemically and metabolically similar to Ca
Ru-106	369 d	0.4	β	
Sb-125	2.77 y	0.03	β, γ	Also produced as an activation product
I-129	1.57×10^7 y	0.6	γ	Volatile
Tc-99m	6.02 h	6.2	β, γ	Volatile; extensively used in nuclear medicine
Cs-134	2.06 y	0.03	β, γ	Semi-volatile; more significant amounts produced by neutron absorption reactions
Cs-137	30.1 y	6.2	β, γ	Semi-volatile
Ce-144	284 d	5.4	β, γ	
<i>Activation Products</i>				
H-3	12.33 y	-	β	Also produced by cosmogenic neutrons; significant releases from CANDU power plants
C-14	5730 y	-	β	Also produced by cosmogenic neutrons
Mn-54	313 d	-	γ	Metabolically important element; multiple oxidation states
Fe-55	2.7 y	-	EC	Metabolically important element
Co-60	5.27 y	-	β, γ	
Sr-89	50.5 d	-	β	Chemically and metabolically similar to Ca
Pu-238	87.7 y	-	α	Metabolically extremely toxic element
Pu-239, Pu-240	2.4×10^4 y, 6.5×10^3 y	-	α, α	Metabolically extremely toxic element
Am-241	432 y	-	α, γ	
<i>Naturally Occurring</i>				
U(U-238, U-235)	4.5×10^9 y, 7.1×10^8 y	-	α, α	
Th(Th-232, Th-230, Th-228)	1.4×10^{10} y, 8.0×10^4 y, 1.9 y	-	α, α, α	
Ra-226	1620 y	-	α	Chemically and metabolically similar to Ca
Ra-228	5.8 y	-	β, γ	Chemically and metabolically similar to Ca
Pb-210	22.3 y	-	β, γ	
Po-210	138 d	-	α	

*Compiled from *The Gamma Rays of the Radionuclides. Tables for Applied Gamma Ray Spectrometry*, Erdtmann and Soyka (1979).
 †y = years, d = days.

Radionuclide Monitoring

The Inland Waters Directorate (IWD) is responsible for providing data and information relating to the presence of toxic chemicals in the aquatic environment. Radionuclides are known to exist in surface waters, but the available data are scarce. Other agencies such as provincial departments of the environment and Health and Welfare Canada are carrying out radiological monitoring programs to assess the levels of radionuclides in potable waters across Canada. In such cases, IWD makes arrangements to obtain the data from those agencies. The National Radionuclides Monitoring Program (NRMP), conducted jointly by the Water Quality Branch and the National Water Research Institute (NWRI) of Environment Canada since 1980, does not duplicate those efforts.

Objectives

The objectives of this report are to summarize the available data, identify radiological water quality trends, and assess the environmental significance of the results obtained to date. Based on the analysis of the results and a

review of the objectives of the National Radionuclides Monitoring Program, the report will also recommend changes, if necessary. The data collection program results reported are for the period 1981 to 1984 inclusive.

DATA COLLECTION PROGRAM

Sampling Locations

The sampling locations were chosen for their potential for containing higher than background levels of radionuclides in the aquatic environment due to nuclear fuel cycle operations, or due to coal and uranium mineralization/mining activities. In addition, a few baseline sampling stations were included in the sampling program (Water Quality Branch, 1980). Areas of possible concern that were being monitored adequately by other agencies were not included in the program. The sampling stations monitored by the Water Quality Branch are listed in Table 2, and their approximate locations are shown in Figure 3. Generally, sampling near point sources was avoided, as most of these locations are adequately monitored by other agencies.

Table 2. Sampling Stations Monitored by the Water Quality Branch for the National Radionuclides Monitoring Program

Sampling location*	NAQUADAT number	Sampling station		Reasons for monitoring
		Northern longitude	Western latitude	
<i>Surface Water Sampling</i>				
1. Ranger Bight, Lab.	00NF 03NG 0004	55°04'54"	59°12'29"	Potential uranium mineralization/exploration
2. Annapolis River, N.S.	00NS 01DC 0001	44°57'14"	65°00'10"	Baseline station
3. McAskill Brook, N.S.	00NS 01FJ 0021	46°09'06"	59°57'47"	Coal mineralization/mining
4. Moose Creek, N.B.	02NB 01AQ 0001	45°07'04"	66°22'44"	Nuclear power generation
5. Petitcodiac River, N.B.	02NB 01BU 0002	46°04'28"	64°47'03"	Uranium fuel assembly plant
6. Niagara River at Niagara-on-the-Lake, Ont.	000N 02HA 0019	43°15'20"	79°03'20"	Industrial waste dumps; possibility of radionuclide leaching into the river
7. Niagara River at Fort Erie, Ont.	—	42°52'00"	78°55'00"	Nuclear fuel reprocessing plant wastes from West Valley, New York
8. St. Lawrence River at Wolfe Island, Ont.	000N 02MA 0030	44°12'00"	76°13'59"	Nuclear-related activities
9. Souris River near Coulter, Man.	00MA 05NF 0001	49°05'27"	100°56'51"	Potential coal mining
10. Red River, Man.	00MA 05OC 0001	49°00'29"	97°12'38"	Industrial activities
11. Saskatchewan River at Man.-Sask. Boundary	00MA 05KH 0001	53°50'30"	101°20'03"	Several coal belts and thermal plants drain into the river
12. Poplar River East, Sask.	00SA 11AE 0008	49°00'00"	105°24'33"	Coal mining area
13. Baker Lake, N.W.T.	01NW 06MA 0001	64°19'09"	96°01'49"	Uranium mineralization/exploration
<i>Sediment Sampling</i>				
14. Ottawa River/Bissett Creek, Ont.	—	46°10'00"	78°05'00"	Baseline station
15. Ottawa River/Petawawa, Ont.	—	45°52'00"	77°17'00"	Nuclear power generation/nuclear laboratories (CRNJ.) effluent discharges

*Sampling locations are indicated in Figure 3.

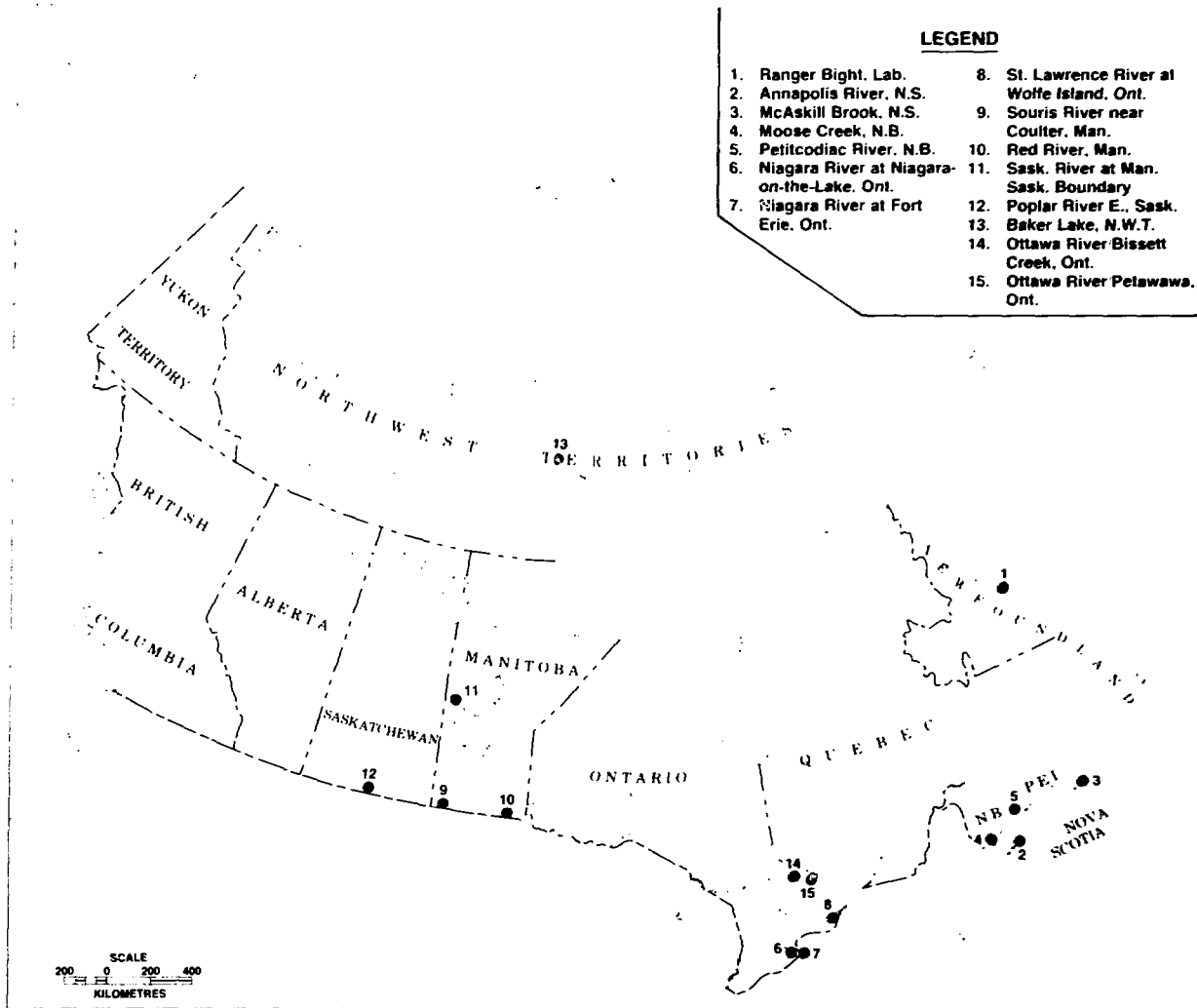


Figure 3. Map showing approximate locations of sampling stations monitored across Canada.

Sample Collection

Water samples were collected by WQB personnel. Four-litre samples were collected monthly in polyethylene containers and acidified with 8 mL concentrated hydrochloric acid as preservative. The 12 monthly samples (January to December) were composited and a representative 5-L sample was sent to the NWRI annually for radionuclide analyses. However, for the St. Lawrence (at Wolfe Island) and Niagara rivers (Niagara-on-the-Lake) samples were composited and analyzed quarterly for three years (1982 to 1984). In addition, water samples from the Niagara River at Fort Erie were also analyzed quarterly for

the year 1984. Sediment samples from the Ottawa River at two locations, up and downstream from nuclear facilities, were also collected by the WQB Ontario Region as part of a bottom sediment survey of the river in 1982.

Analytical Methods

Water samples were generally analyzed for tritium, antimony-125, cesium-137, radium-226, and total uranium. The Ottawa River sediments were analyzed for lead-210, cesium-137, radium-226, and cobalt-60. Methods used to analyze the samples are briefly described below.

Water Samples

Samples were analyzed for H-3 immediately upon arrival at NWRI by adding 8-17 mL of PCS, an Amersham scintillation cocktail, to a polyethylene counting vial and counting for 200 min in a Searle Mark II liquid scintillation counter at 12°C. The counter was standardized with an Amersham tritiated water standard and the pulse-height analyzer window adjusted to contain only the central 50% of the H-3 energy peak for minimum background contribution. Background was determined using a near-zero level H-3 water sample.

Uranium was determined on 5-mL aliquots using a Scintrex laser-fluorescence analyzer. Additions of standard amounts of uranium to the water/fluor mixture were made to determine quenching effects. The uranium determined in this way has been defined as "total U" in this report and essentially represents the isotopic composition of natural uranium.

Samples containing 2-50 L water were evaporated to a final volume of 40 mL to determine the gamma-emitting radionuclides Cs-137 and Sb-125 (Durham, 1974; Durham and Joshi, 1984). Concentrated nitric acid was added to the sample in small amounts when the volume reached about 200 mL in order to destroy organic materials. Experiments using respective radiotracers have established that this pre-concentration procedure gives quantitative recoveries. The gamma-ray spectra of the final 40-mL samples, in polyethylene bottles, were measured with two liquid nitrogen-cooled hyperpure germanium detectors in chambers shielded with 10 cm of lead. The detectors, in coaxial configuration, were interfaced to a Nuclear Data 6620 nuclear spectrometer set up to count samples for 2.5×10^5 s with 2048 channels of spectral data at 1 keV per channel for each detector. The detectors were standardized using Amersham Canada standardized radionuclides in the same 40-mL geometry. The count rates of the photopeaks were converted to disintegration rates using detector efficiency data and the gamma-emission probabilities of the radionuclides. Figure 4 shows the typical gamma-ray spectrum obtained for a 1984 Niagara River water sample.

The concentrations of Ra-226 in water samples were derived using the standard radon-emanation technique as described elsewhere by Durham and Joshi (1980a), except that the Ba/RaSO₄ step was replaced by direct dilution of the above 40-mL sample with doubly-distilled deionized water to the desired level. Unsatisfactory results were obtained when attempts were made to measure Ra-226 by gamma-ray spectrometry using the 186- or 352-keV emissions of this radionuclide.

Sediment Samples

The analyses of the two Ottawa River sediment samples for Pb-210, Ra-226, Cs-137 and Co-60 were also done by gamma-ray spectrometry. The Pb-210 and Ra-226 contents of the samples were measured on intrinsic germanium detectors in planar configuration using photopeaks at 46.5 (Pb-210), and 186 and 352 (Ra-226) keV. The concentrations of Cs-137 and Co-60 were derived from count rates measured on an intrinsic germanium detector in coaxial configuration using the photopeaks at 662 (Cs-137), and 1173 and 1332 (Co-60) keV. Experiments using epoxy-layered samples revealed that emanation of Rn-222 (and the resulting change in geometry) from non-layered samples in standard counting vials is negligible and hence the 352-keV photopeak can be used for measuring Ra-226 in solid samples. Furthermore, the results from the 186-keV photopeak were identical with those afforded by the 352-keV peak. The efficiencies of the detectors were determined at four different thicknesses as described by Joshi (1985).

Quality Assurance

Quality control/quality assurance is an important factor for obtaining reliable data that can be interpreted meaningfully. For this particular program, field staff followed the general procedures as suggested for sample collection and shipping in the "Sampling for Water Quality" manual (Environment Canada, 1983). For sample preservation, the staff were appropriately instructed.

The quality of the analytical data was regularly assessed through continuous participation in intercomparison programs organized by the U.S. Environmental Protection Agency and through selective participation in similar programs organized by the International Atomic Energy Agency. Internal quality control was achieved by analyzing certified reference materials procured from CANMET-EMR (Canada Centre for Mineral and Energy Technology - Energy, Mines and Resources Canada), the U.S. National Bureau of Standards, the U.S. Environmental Protection Agency, and the International Atomic Energy Agency.

RESULTS

Under the National Radionuclides Monitoring Program, radionuclides in some surface waters were monitored across Canada during the period 1981 to 1984. The sampling stations were selected essentially based on potential radiological impacts on water quality due to uranium and coal mineralization, emissions/discharges from nuclear power stations, industrial effluent discharges including

COUNTS PER CHANNEL

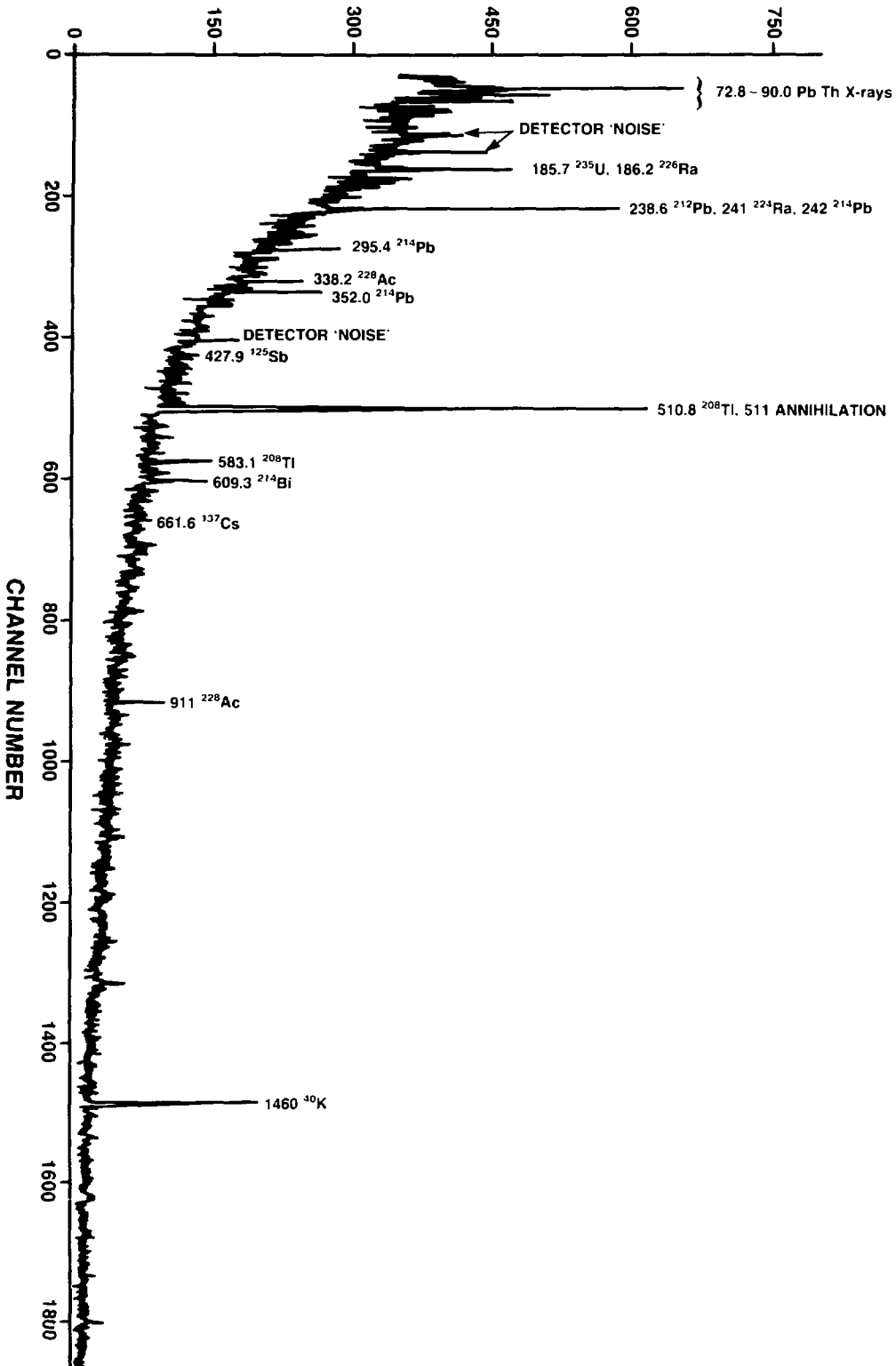


Figure 4. Gamma-ray spectrum of a 1984 Niagara River water sample showing prominent photopeaks with their energies (keV).

those from uranium mining and milling, and coal mining (Table 2).

The sampling stations monitored are listed in Table 3. The number of stations monitored varied during each year; they were 10, 11, 11, and 13 for the years 1981, 1982, 1983 and 1984, respectively. During 1981, the annual water composite samples were analyzed for radium-226 (Ra-226), tritium (H-3), and total uranium (U). In subsequent years, two more radionuclides, cesium-137 (Cs-137) and antimony-125 (Sb-125), were added. Limited resources prevented inclusion of Sr-90 in these measurements, although it is well recognized that of all the fallout radionuclides, Sr-90 provides the most radiation dose (Durham and Joshi, 1984).

The detailed analytical data for the annual composite water samples are presented in Tables A-1 to A-4 in the Appendix. Errors shown with individual measurements are the statistical errors of measurement including those in the background. All the available data for the monitoring years are summarized in Table 4. Herein the individual data are means of the four-year data with their standard deviations. To study short-term temporal variations, some samples from the Ontario locations were analyzed quarterly; the data for 1982 to 1984 are presented in Table 5.

For sediments, only two locations were sampled along the Ottawa River. The intersection of the Ottawa River at Bissett Creek is located upstream from the water intake/discharge for the nuclear power station (NPD), whereas the intersection of the Ottawa River at Petawawa is located below the AECL nuclear laboratories at Chalk River, Ontario. The former station was chosen to represent base-

line (background) conditions in the Ottawa River, and the latter station should reflect the impact of discharges from the two nuclear facilities. The sediment data from the two Ottawa River locations in Ontario are presented in Table 6.

Table 3. Sampling Stations Monitored Each Year during Sampling Period, 1981 to 1984

Sampling location*	Year of monitoring			
	1981	1982	1983	1984
<i>Surface Water Sampling</i>				
1. Ranger Bight, Lab.		x	x	x
2. Annapolis River, N.S.	x	x	x	x
3. McAskill Brook, N.S.	x	x	x	x
4. Moose Creek, N.B.	x	x	x	x
5. Petitcodiac River, N.B.	x	x	x	x
6. Niagara River at Niagara-on-the-Lake, Ont.	x	x	x	x
7. Niagara River at Fort Erie, Ont.			x	x
8. St. Lawrence River at Wolfe Island, Ont.	x	x	x	x
9. Souris River near Coulter, Man.	x	x	x	x
10. Red River, Man.			x	x
11. Saskatchewan River at Man.-Sask. Boundary	x	x		x
12. Poplar River East, Sask.	x	x		x
13. Baker Lake, N.W.T.	x	x	x	x
<i>Sediment Sampling</i>				
14. Ottawa River/Bissett Creek, Ont.		x		
15. Ottawa River/Petawawa, Ont.		x		

*Sampling locations are indicated in Figure 3.

Table 4. Average Radionuclide Concentration in Surface Waters, 1981 to 1984

Sampling location*	Average radionuclide concentration†				
	Ra-226 (mBq/L)	Cs-137 (mBq/L)	Sb-125 (mBq/L)	Tritium (Bq/L)	Total U (µg/L)
1. Ranger Bight, Lab.	4.44±1.11	<13.69	<7.78	5.95±2.34	0.70±0.08
2. Annapolis River, N.S.	1.48±0.30	<3.97	<8.98	6.02±1.49	0.43±0.05
3. McAskill Brook, N.S.	1.48±0.60	<6.66	15.56±9.45	4.70±1.15	0.30±0.04
4. Moose Creek, N.B.	1.66±0.64	<3.39	<8.88	6.81±1.78	1.37±0.26
5. Petitcodiac River, N.B.	12.67±10.14	<4.44	<7.17	7.88±1.73	2.24±1.91
6. Niagara River at Niagara-on-the-Lake, Ont.	2.19±0.86	0.70±0.32	<1.81	8.00±0.88	0.48±0.08
7. Niagara River at Fort Erie, Ont.	1.66±0.26	<1.00	1.05±0.40	5.12±2.06	0.28±0.17
8. St. Lawrence River at Wolfe Island, Ont.	1.43±0.53	1.52±0.33	<2.40	8.56±1.67	0.57±0.27
9. Souris River near Coulter, Man.	2.13±1.22	<4.46	<6.93	7.64±5.46	2.39±1.14
10. Red River, Man.	3.88±3.92	5.50±4.33	<7.52	<6.33	4.74±3.05
11. Saskatchewan River at Man.-Sask. Boundary	5.92±1.70	<4.63	<8.35	<9.92	0.99±0.07
12. Poplar River East, Sask.	7.03±0.37	<3.50	<7.24	<6.48	2.84±0.76
13. Baker Lake, N.W.T.	1.57±0.70	4.22±1.77	<10.19	12.24±4.68	0.16±0.08
Mean	3.66	—	—	—	1.35

*Sampling locations are indicated in Figure 3.

†The values reported for each radionuclide are means of the available data with their standard deviations.

Table 5. Quarterly Analyses of Surface Water Samples from Ontario

Year	Sampling location*		Radionuclide concentration†				
			Ra-226 (mBq/L)	Cs-137 (mBq/L)	Sb-125 (mBq/L)	Tritium (Bq/L)	Total U (µg/L)
1982	Niagara River at Niagara-on-the-Lake	1Q‡	2.33±0.44	1.27±0.78	<1.81	6.70±2.37	0.40±0.02
		2Q	3.77±0.67	-	-	8.18±3.44	0.43±0.07
		3Q	1.74±0.07	0.52±0.18	0.37±0.33	12.28±3.44	0.54±0.08
		4Q	1.52±0.07	0.96±0.18	<0.30	7.55±1.52	0.65±0.10
		Mean	2.34±1.02	0.92±0.38	<1.81	8.68±2.48	0.50±0.11
	St. Lawrence River at Wolfe Island	1Q	0.74±0.37	2.04±0.81	<2.40	10.14±2.37	-
		2Q	1.48±0.74	2.74±0.81	2.04±1.78	9.47±3.66	0.40±0.66
		3Q	1.48±0.15	0.85±0.67	<1.70	10.32±3.44	2.11±0.32
		4Q	1.55±0.22	1.41±0.63	<1.33	11.84±1.52	0.42±0.06
		Mean	1.31±0.38	1.76±0.81	<2.40	10.44±1.00	0.98±0.98
1983	Niagara River at Niagara-on-the-Lake	1Q	0.63±0.04	0.28±0.25	<0.47	9.25±2.81	0.60±0.09
		2Q	1.11±0.07	<0.23	<0.61	<4.11	0.62±0.09
		3Q	2.22±0.07	0.36±0.23	1.16±0.77	9.69±2.81	0.60±0.09
		4Q	1.48±0.07	0.77±0.22	<0.74	6.03±2.37	0.50±0.08
		Mean	1.36±0.67	0.47±0.26	<1.16	8.32±2.00	0.58±0.05
	St. Lawrence River at Wolfe Island	1Q	1.18±0.15	<0.74	<1.50	8.62±2.81	0.50±0.08
		2Q	0.78±0.07	1.36±0.55	<1.12	<4.11	0.44±0.07
		3Q	0.93±0.07	1.45±0.57	<1.45	7.55±2.81	0.42±0.06
		4Q	1.48±0.11	1.05±0.76	<1.09	5.60±2.37	0.43±0.06
		Mean	1.09±0.31	1.29±0.21	<1.50	7.26±1.53	0.45±0.04
1984	Niagara River at Niagara-on-the-Lake	1Q	2.22±0.07	0.53±0.34	1.28±0.84	<2.07	0.42±0.06
		2Q	-	-	-	7.44±1.90	0.47±0.07
		3Q	1.85±0.07	<0.24	<0.80	5.99±1.90	0.42±0.06
		4Q	1.11±0.07	0.73±0.31	<0.58	7.56±1.97	0.48±0.07
		Mean	1.73±0.56	<0.73	<1.28	7.00±0.87	0.45±0.03
	Niagara River at Fort Erie	1Q	1.48±0.07	<0.29	<0.59	7.73±2.15	0.39±0.06
		2Q	1.48±0.07	<0.25	<0.77	4.81±1.89	0.50±0.08
		3Q	1.85±0.07	<0.34	<0.63	8.07±1.90	0.38±0.06
		4Q	1.11±0.04	0.50±0.29	<0.68	5.70±1.96	0.33±0.05
		Mean	1.48±0.30	<0.50	<0.77	6.58±1.58	0.40±0.07
St. Lawrence River at Wolfe Island	1Q	1.11±0.07	<0.50	<1.17	7.66±2.15	0.50±0.08	
	2Q	1.11±0.07	<0.48	<0.99	8.19±1.90	0.43±0.06	
	3Q	1.11±0.07	1.01±0.50	<1.02	9.87±1.91	0.38±0.06	
	4Q	1.11±0.07	<0.47	<1.16	6.16±1.97	0.43±0.06	
	Mean	1.11±0.07	<1.01	<1.17	7.97±1.53	0.43±0.05	

* Sampling locations are indicated in Figure 3.

† The errors reported with quarterly measurements are the statistical errors of the individual measurements including those in the background. Errors reported with quarterly means, however, are standard deviations.

‡ Q represents quarter of the year.

Table 6. Radionuclide Concentration in Sediments of the Ottawa River, 1982

Sampling location*	Radionuclide concentration (Bq/g)†				
	Pb-210	Ra-226	Cs-137	Co-60 (1173 keV)	Co-60 (1332 keV)
Ottawa River/Bissett Creek, Ont.	0.56±0.02	0.04±0.01	0.39±0.01	<0.01	<0.01
Ottawa River/Petawawa, Ont.	0.57±0.02	0.04±0.01	1.32±0.01	0.21±0.01	0.21±0.01

* Sampling locations are indicated in Figure 3.

† The errors reported with each measurement are the statistical errors of the individual measurements including those in the background.

Generally, the radioactivity levels due to the presence of radionuclides, particularly Ra-226 and total U, in surface waters were slightly higher in samples collected during 1983 than in any other year (Appendix Tables A-1 to A-4, and Table 4). The levels of total uranium and Ra-226 were all above detection limit, whereas those for the fission products, especially Cs-137 and Sb-125, were mostly below the detection limit. As is evident from these data, the detection limits have been variable for different samples even for the same radionuclide. This is due to fluctuating levels of detector background which are known to affect the detection limits.

For total U, annual mean values for all the sampling locations varied from 0.90 to 2.05 $\mu\text{g/L}$. The lowest value, 0.10 $\mu\text{g/L}$, was from the Baker Lake location in 1982 and the highest value, 6.90 $\mu\text{g/L}$, was found in the Red River location in 1983. Annual means for Ra-226 varied from 2.13 to 4.60 mBq/L, the lowest being only 0.74 mBq/L in samples from McAskill Brook and Souris River in 1984, and the highest being 23.68 mBq/L recorded in the Petitcodiac River water in 1983. The highest levels of Cs-137, Sb-125 and tritium were 13.69 mBq/L at Ranger Bight (1982), 22.24 mBq/L at McAskill Brook (1983), and 17.46 Bq/L at Baker Lake (1981), respectively. Mean values for these radionuclides cannot be computed because most of the measurements were below the detection limit.

DISCUSSION

Nature of Radioactivity

Radioactivity in the environment results from the decay processes of individual radionuclides which may originate from natural and man-made sources. The U-238 and Th-232 decay chain radionuclides are the major source of naturally occurring radionuclides (Figs. 1 and 2). The man-made radionuclides primarily result from atmospheric fallout due to nuclear weapons testing and nuclear fuel cycle operations. Table 1 summarizes basic properties of some environmentally important radionuclides.

Unlike the International Joint Commission, which primarily has concentrated its efforts on the measurement of man-made radionuclides in the Great Lakes (Durham, 1974; Durham and Joshi, 1984; Great Lakes Water Quality Board, 1981; 1983), this program included the measurements of uranium and radium that originate primarily from natural sources. The concentrations of these radionuclides, however, may be enhanced due to man's activities. For example, the levels of radium in both water and sediments are known to be elevated in the vicinity of the Serpent River mouth due to uranium mining and milling in the

Elliot Lake district upstream (Hart and McKee, 1985; Great Lakes Water Quality Board, 1983; Meyerhof, 1984). The Serpent River discharges through Serpent Harbour into Lake Huron.

Radionuclide Levels and Trends

Annual means over all sampling stations, wherever computable, and other data suggest that no trends are discernible for levels of radionuclides in surface waters (Appendix Tables A-1 to A-4). In general, no trends would be expected for the radionuclides of natural origin, i.e., total U and Ra-226, since the inputs from natural sources are usually constant. In contrast, the levels of man-made radionuclides (Cs-137, Sb-125 and tritium) were expected to decrease with time as had been found for the Great Lakes (Durham and Joshi, 1984; Tracy and Prantl, 1983). This decrease of fission products in the Great Lakes was primarily attributed to a reduced fallout input as a result of a Nuclear Treaty in 1963 banning the atmospheric testing of nuclear weapons. Some atmospheric testing has been carried out by the Peoples Republic of China and France, however, and the presence of radioactive fallout from these detonations in the Canadian aquatic ecosystems is well documented (Environmental Health Directorate, 1979; Roy *et al.*, 1981). Lack of any obvious trends in Cs-137, Sb-125 and H-3 concentrations may be because most of the data for the Great Lakes studies, the present study encompasses the time period corresponding to minimal atmospheric nuclear explosions.

Cesium-137 and Antimony-125

As mentioned earlier, levels of most of the anthropogenic radionuclides were below the detection limits, especially for Cs-137 and Sb-125 (Table 4). In general, these data are in the same range as has been found for the Great Lakes in the early 1980s (Durham and Joshi, 1981, 1984; Great Lakes Water Quality Board 1981, 1983). Besides the reduced inputs from fallout, both Cs-137 and Sb-125 are quite reactive with the sediments. For example, Tracy and Prantl (1983) estimated that as much as 93% and 96% of the total Cs-137 activity was stored in the sediments of Lakes Superior and Huron, respectively. For Sb-125, the values ranged from 25% to 50%. Thus the levels of both Cs-137 and Sb-125 left in the aqueous phase would be rather small.

Tritium

The levels of tritium in the surface waters were generally above the detection limit (Appendix Tables A-1 to A-4, and Table 4) but essentially in the same range as has recently been found in the open waters of the Great Lakes

(Great Lakes Water Quality Board, 1983). Most of the tritium present in the aqueous environment has originated from nuclear weapons testing in the atmosphere (Health and Welfare Canada, 1979b; U.S. EPA, 1981). Again, since the banning of atmospheric testing in 1963, the tritium levels in the aquatic environment have decreased significantly. Kathren (1984) summarized the available data for the U.S. environmental waters and streams; his conclusion was that the levels in 1983 were an order of magnitude lower than those found in 1963.

Nuclear power generation may also release significant quantities of radionuclides to the immediate environments. Recently, available monitoring data were summarized by the Water Quality Branch (1985b) to compare the levels of radionuclides at Rolphton, which is upstream from the nuclear power generating station (NPD) and AECL nuclear laboratories at Chalk River, and at Pembroke, which is downstream from the two nuclear facilities. Means of the five-year data (1979 to 1983) at Pembroke revealed about a twofold increase in the levels of both Cs-137 and tritium, while those of Sr-90 were unaffected.

Uranium and Radium-226

Radioactivity from natural sources originates primarily from the uranium (U-238 and U-235) and thorium (Th-232) decay chain radionuclides. The average concentration of uranium in the earth's crust is about 1 µg/g (B.C. Royal Commission, 1980; Cothorn and Lappenbusch, 1983), whereas that of thorium is somewhat higher. Hess *et al.* (1985) estimated an average Th/U activity ranging from 1.2 to 1.5; hence in the absence of enrichment or depletion processes, the levels of radionuclides from the Th-232 decay chain should be higher than those from the U-238 decay series. However, since thorium is quite insoluble, this radionuclide is expected to be found at very low concentrations in water.

The B.C. Royal Commission (1980) estimated that uranium concentration of most natural surface waters is of the order of about 0.1 µg/L (ppb). Surface waters in contact with uranium mineralization may have a concentration as high as 1 to 5 µg/L. In exceptional cases, the concentration may be as high as 50 µg/L when the water is in contact with uranium ore deposits.

As far as natural radioactivity in the aqueous environment is concerned, uranium and radium-226 are considered most important. Natural uranium is mainly composed of three isotopes: 99.27% of U-238, 0.72% of U-235, and 0.006% of U-234 (Cothorn and Lappenbusch, 1983). At equilibrium, 1 µg of natural uranium has an activity of 24.8 mBq (0.67 pCi).

Average concentrations for total U and Ra-226 (four-year data) are shown in Table 4 along with other radionuclides. Average concentrations of total U have ranged from 0.16 µg/L in the Baker Lake location to 4.74 µg/L in the Red River waters, with an overall mean of 1.35 µg/L. This range is essentially within the range reported for waters in contact with uranium mineralization (1 to 5 µg/L, B.C. Royal Commission, 1980). Most of the surface waters drain either uranium or coal mineralization areas (Table 2); hence the ranges measured were expected. Yet the lowest levels measured in the Baker Lake location (0.16 µg/L) are rather unexpected because it does drain a uranium-rich basin. Coal mineralizations may contain up to 1000 µg/g of U (B.C. Royal Commission, 1980).

Cothorn and Lappenbusch (1983) reviewed the literature relating to U occurrence in the natural waters of the world, especially in relation to that in the U.S.A. They quote a range from 0.02 µg/L in the St. Lawrence River (Canada) to 1.6 µg/L in the Ganges-Bramaputra rivers in India. For surface waters in the United States, they give a value of 1 pCi/L (1.5 µg/L); this value is very close to that reported in Table 4 for Canadian surface waters, i.e. 1.35 µg/L.

For the Great Lakes, the uranium concentrations were somewhat lower, ranging only from 0.08 to 0.59 µg/L during 1981 and 1982 (Great Lakes Water Quality Board, 1983). The radiological water surveys carried out in British Columbia during 1979 to 1981, however, revealed quite high levels of uranium in some surface waters and even higher levels in ground/well waters (B.C. Ministries of Energy, Mines and Petroleum Resources, and of Health, 1980, 1981). Generally, the high values approaching/exceeding 10 µg/L were recorded in uranium mineralization areas of Summerland and Nelson.

Temporal Variations

To study temporal variations, samples were analyzed quarterly, instead of annual composites; the data are presented in Table 5. The standard deviations reported alongside quarterly means are usually much less than 50% of the mean which is not high, considering that most levels of the measured radionuclides are barely above the detection limits.

For the radiological water surveys carried out in British Columbia, most samples were analyzed monthly (B.C. Ministries of Energy, Mines and Petroleum Resources, and of Health, 1980). The reported data reveal cyclic variations; generally, February–April values for uranium were higher than those for May–June values (their Table 14). The

authors postulated that perhaps the water flow data that were not available would have explained such variations. For the short-term variations (daily-weekly), the authors concluded that "temporal compositional variations appear to be slight."

Thus, in view of the results available to date, the composite water samples being analyzed annually should be considered adequate at this time.

Radionuclide Content of Sediments

Sediments can serve as a potential sink/source for reactive pollutants including radionuclides. The magnitude of radioactivity in the aqueous and solid phases depends on the relative concentration of radionuclides in the respective phases; thus transference from one phase to another will occur depending upon concentration gradients (Tracy and Prantl, 1983). In flowing waters, the transport of radionuclides may take place in true solution, or by attachment to fine particulates (Martin and Meybeck, 1979). For the St. Lawrence River, Lupien and Grondin (1984) estimated that over 50% of the natural radionuclides measured in the aqueous phase were associated with or attached to the suspended solids.

Concentrations of radionuclides in the sediments are shown in Table 6. The levels of the naturally occurring Pb-210 and Ra-226 are essentially in the same range as reported for Lake Huron by Durham and Joshi (1980b). The Cs-137 levels also appear to be in the range measured for Lake Huron sediments; on closer inspection, however, the levels are higher in the Ottawa River sediments, since the measured levels for Lake Huron bottom sediments, in fact, represent nuclear weapons fallout accumulated over several years. The presence of Co-60 in Ottawa River sediments also is evidence of the occurrence of radionuclides from Chalk River in the study area.

The locations of the sampling stations are shown in Table 2. The Bissett Creek intersection of the Ottawa River is located upstream from two nuclear facilities, the nuclear power generating station (NPD) and AECL nuclear laboratories at Chalk River, Ontario. The other location, near Petawawa, is located downstream from the two nuclear facilities; hence the radionuclide concentrations differential should reveal the impact of the two nuclear facilities on sediment quality.

The data in Table 6 clearly reveal the impact of the two nuclear facilities on the radionuclide concentrations in the sediments. Cesium-137 and Co-60 were significantly elevated, whereas the levels of Pb-210 and Ra-226 were unaffected. The Ottawa River water quality data, in a review to assess the radiological impact of these two nuclear

facilities, indicated that Cs-137 was elevated approximately twofold downstream at Pembroke compared with background levels at Rolphton (Water Quality Branch, 1985b). Thus, it appears that Cs-137 and perhaps Co-60 have migrated from the aqueous phase into the sediments. Contrary to this phenomenon, Tracy and Prantl (1983) concluded that Cs-137 is now re-entering water from the sediments in Lakes Superior and Huron due to reduced fallout input in the recent years. The sediment data from other surface water bodies surveyed are not available.

Radiological Health Impacts

Drinking water can serve as a significant pathway for entry of radionuclides into biota and humans (Health and Welfare Canada, 1979b; U.S. EPA, 1981). Specifically, for uranium intake in the U.S.A., Cothorn and Lappenbusch (1983) estimated that consumption of water contributed about 85% of the intake, whereas the remaining 15% was contributed by food.

Water Quality Guidelines

To protect the general public from excessive radiation exposure, revised guidelines for the consumption of drinking water were proposed by the Health and Welfare Canada in 1978; the recommended limits are shown in Table 7. The guidelines are based on a linear dose-response relationship as recommended by the International Commission on Radiological Protection (ICRP) in publications 26 and 30 (ICRP, 1977, 1979). Maximum acceptable concentration (MAC) in drinking water has been derived which corresponds to 1% (0.5 mSv) of the ICRP recommended annual occupational dose equivalent limit for continuous exposure, while target concentration (TC) corresponds to only 10% (0.05 mSv) of the MAC. Radionuclide concentrations exceeding the MAC may be tolerated, provided that the duration of the increase is short and that the annual average concentration remains below this limit.

Table 7. Recommended Guidelines for Radionuclide Concentrations in Drinking Water

Radionuclide	Maximum acceptable concentration (Bq/L)	Target concentration (Bq/L)
Cesium-137	50	5
Iodine-131	10	1
Radium-226	1	0.1
Strontium-90	10	1
Tritium	40 000	4000
Uranium (mg/L)	0.02	≤0.001

Source: *Guidelines for Canadian Drinking Water Quality 1978*, Health and Welfare Canada (1979a).

Average radionuclide concentrations of both naturally occurring (total U and Ra-226) and man-made radionuclides

(Cs-137, Sb-125 and H-3) are shown in Table 4. As mentioned earlier, the levels of man-made radionuclides were generally quite low. The levels of both tritium and Cs-137 were much below the recommended target concentration of 4000 and 5 Bq/L, respectively. The levels of Sb-125 are quite low as well, but no recommended guidelines exist either for Canada or the United States (Health and Welfare Canada, 1979a; U.S. EPA, 1981).

All the levels of naturally occurring Ra-226 are below the target concentration of 100 mBq/L (Table 7). Similar conclusions were reached by Monenco Consultants Limited (1981), who surveyed 99 municipal drinking water supplies in Alberta; their Ra-226 concentrations ranged from 3.7 to 11.1 mBq/L (0.1 to 0.3 pCi/L). However, this is not the case for total uranium; five individual sampling stations and the overall average of 1.35 µg/L exceed the target concentration of 1.0 µg/L. In contrast, none of the Great Lakes concentrations of uranium exceeded the TC during 1981 and 1982 (Great Lakes Water Quality Board, 1983). However, some of the water samples in British Columbia even exceeded the MAC of 20 µg/L during certain monthly sampling periods (B.C. Ministries of Energy, Mines and Petroleum Resources, and of Health, 1980; 1981).

Radiological Dose Commitment

Annual dose commitment¹ provides a quantitative assessment of the radiological hazard posed by the ingestion of waters containing radionuclides. In this study, doses

¹The basis for radiological dose commitment calculation is the annual limit of intake (ALI) as recommended in ICRP 30. The ALI is the quantity of a given radionuclide which, when injected or inhaled, will result in an uptake that will yield a committed dose equivalent of 5 rem (0.05 Sv) over the subsequent 50-year period.

were calculated using the data in Table 4. The methodology used was similar to that used by the International Joint Commission (Great Lakes Water Quality Board, 1983). For the purposes of calculation, a daily ingestion of 2.2 L (803 L/y) was assumed. The basis of dose conversion from radionuclide concentration is the annual limit of intake (ALI), which gives an effective dose equivalent of 5 rem (0.05 Sv). The ALI and concentration-to-dose conversion factors are shown in Appendix Table A-5. Except for total U, all other radionuclide data in Table 3 are in the SI units of radioactivity (becquerel). Therefore, uranium data were converted into radioactivity units assuming an equilibrium among the three isotopes—U-238, U-235, U-234—before dose calculation (Appendix Table A-6).

Radiological doses for the whole body are shown in Table 8. As is apparent, the overall committed effective dose equivalent from naturally occurring radionuclides (total U and Ra-226) was much higher than that from the artificially produced radionuclides (Cs-137, Sb-125 and H-3). Thus, dose contribution from individual radionuclides may be summarized as follows:

$$\text{Total U} > \text{Ra-226} > \text{Tritium} > \text{Cs-137} > \text{Sb-125}$$

The highest dose commitment would occur by the ingestion of water from the Red River, whereas the water from Baker Lake would give the lowest effective dose equivalent.

Unlike the Great Lakes waters, most of the dose commitment was from naturally occurring total U and Ra-226 (Durham and Joshi, 1984; Tracy and Prantl, 1983). In the former case, most of the dose was contributed by Cs-137 and Sr-90. In both cases, however, the dose commitment was less than the objective dose of 1 mrem (10 µSv) as stipulated in the Great Lakes Water Quality Agreement of 1978 (International Joint Commission, 1978).

Table 8. Whole Body Radiological Dose Commitment from the Use of Surface Water as a Source of Drinking Water

Sampling location*	Committed effective dose equivalent (µSv)				
	Ra-226	Cs-137	Sb-125	Tritium	Total U
1. Ranger Bight, Lab.	1.05	<0.15	<0.0041	0.0821	1.12
2. Annapolis River, N.S.	0.35	<0.04	<0.0047	0.0831	0.69
3. McAskill Brook, N.S.	0.35	<0.07	0.0082	0.0649	0.48
4. Moose Creek, N.B.	0.39	<0.04	<0.0047	0.0940	2.20
5. Petitcodiac River, N.B.	2.99	<0.05	<0.0039	0.1087	3.59
6. Niagara River at Niagara-on-the-Lake, Ont.	0.52	<0.01	<0.0010	0.1104	0.77
7. Niagara River at Fort Erie, Ont.	0.39	<0.01	<0.0006	0.0707	0.45
8. St. Lawrence River at Wolfe Island, Ont.	0.34	0.02	<0.0013	0.1181	0.91
9. Souris River near Coulter, Man.	0.50	<0.05	<0.0037	0.0682	3.83
10. Red River, Man.	1.31	0.06	<0.0040	<0.0874	7.60
11. Saskatchewan River at Man.-Sask. Boundary	1.40	<0.05	<0.0044	<0.1369	1.59
12. Poplar River East, Sask.	1.66	<0.04	<0.0038	<0.0894	4.55
13. Baker Lake, N.W.T.	0.37	0.05	<0.0054	0.1689	0.26

*Sampling locations are indicated in Figure 3.

Committed effective dose equivalent from drinking surface waters may be compared with other concepts. Recently, the Atomic Energy Control Board (1985) proposed a *de minimis*, or trivial, dose concept, which states that certain contaminated materials be exempt from licensing if the annual dose commitment is less than 50 μ Sv. Incidentally, this dose commitment is the same as that which would accrue from the continuous consumption of water at "target concentration" (Health and Welfare Canada, 1979a). Thus, these doses are much higher than those occurring from the consumption of surface waters (Table 8).

Radiological doses may be related to expected cancer risks. The Atomic Energy Control Board (1985) proposed an average risk conversion factor of 2×10^{-2} per sievert for both sexes and all ages. Using this conversion factor, the Red River water with the highest dose would give a risk of much less than 10^{-6} per year; this level of risk is generally considered insignificant (Health and Welfare Canada, 1979b; U.S. EPA, 1981).

U-238/Ra-226 Activity Ratios

Total uranium is essentially composed of uranium-238 (99.27% by weight). Radium-226 is a daughter product of the U-238 decay chain series. Uranium-238 and its daughter products are generally assumed to be in secular equilibrium; in weathered geologic materials, however, usually disequilibrium exists (B.C. Royal Commission, 1980). At secular equilibrium, the activities of both radionuclides should be equal; hence deviation from an activity ratio of unity will reveal the extent of disequilibrium and relative mobilities of the two radionuclides.

From total uranium (Table 4), activities of U-238 have been calculated (Appendix Table A-6); U-238 and

Ra-226 activities, and their activity ratios are shown in Table 9. The activity ratios ranged from 1.25 in the Baker Lake to slightly over 10 in Moose Creek waters, with an overall average of 3.04. From Table 8, activity ratios for the Souris River and the Red River have been omitted; they were rather large, 13.72 and 14.94, respectively. These activity ratios simply suggest that U-238 has been much more mobile than Ra-226.

Veska (1983) studied the migration of radionuclides from a uranium waste rock pile near Bancroft, Ontario. He observed that U-238 had generally migrated much farther away from the source than Ra-226 (his Figure 5.23). Similar conclusions were reached by King *et al.* (1982) and Krishnaswami *et al.* (1982), who studied the relative mobilities of the two radionuclides.

In another study, Lupien and Grondin (1984) investigated the levels of natural radionuclides in the St. Lawrence River. From their Table 1, U-238/Ra-226 activity ratios were calculated; they ranged from 2.5 to 24.0. In addition, their K_d values for U-238 and Ra-226 (their Table 2) would also suggest that U-238 is much more mobile relative to Ra-226.

The mechanisms proposed for the relative mobilities of the two radionuclides are the adsorption of ionic radium onto solid surfaces mostly via ion exchange, while anionic complexes of uranium species with carbonate and hydroxyl ions are quite stable and mobile in the aqueous environment (King *et al.*, 1982; Krishnaswami *et al.*, 1982; Veska, 1983).

Regression analyses were performed to determine whether Ra-226 concentration in surface waters would be predictable from total uranium concentration. In these

Table 9. Average Uranium and Radium Concentrations, and U-238/Ra-226 Activity Ratios in Surface Waters, 1981 to 1984

Sampling location*	Radionuclide concentration (mBq/L)		U-238/Ra-226 activity ratio
	U-238†	Ra-226	
Ranger Bight, Lab.	8.56	4.44	1.93
Annapolis River, N.S.	5.26	1.48	3.55
McAskill Brook, N.S.	3.67	1.48	2.48
Moose Creek, N.B.	16.76	1.66	10.10
Petitcodiac River, N.B.	27.40	12.67	2.16
Niagara River at Niagara-on-the-Lake, Ont.	5.87	2.19	2.68
Niagara River at Fort Erie, Ont.	3.42	1.66	2.06
St. Lawrence at Wolfe Island, Ont.	6.97	1.43	4.87
Saskatchewan River at Man.-Sask. Boundary	12.11	5.92	2.05
Poplar River East, Sask.	34.74	7.03	4.94
Baker Lake, N.W.T.	1.96	1.57	1.25
Mean	11.50	3.78	3.04

*Sampling locations are indicated in Figure 3.

†Concentration of U-238 was calculated from total uranium using a concentration factor of 0.9927.

analyses, total U was treated as an independent variable, while Ra-226 was considered a dependent variable. Appendix Figure A-1 shows a curvilinear relationship when all values were used; an insignificant correlation coefficient (r) value of only 0.542 was obtained. Yet when the two extreme values, i.e. Souris River and Red River, were excluded, a highly significant correlation coefficient of 0.776^{**} ($n = 11$, $P < 0.01$) was obtained (Appendix Fig. A-2). Nonetheless, when the data were plotted through the origin, a somewhat lower but highly significant correlation coefficient ($r = 0.761^{**}$) was obtained, as shown in Figure 5. Thus, it may be concluded that for most water samples, total U and Ra-226 levels are related, and the concentration of either one can be estimated from the concentration of the other radionuclide.

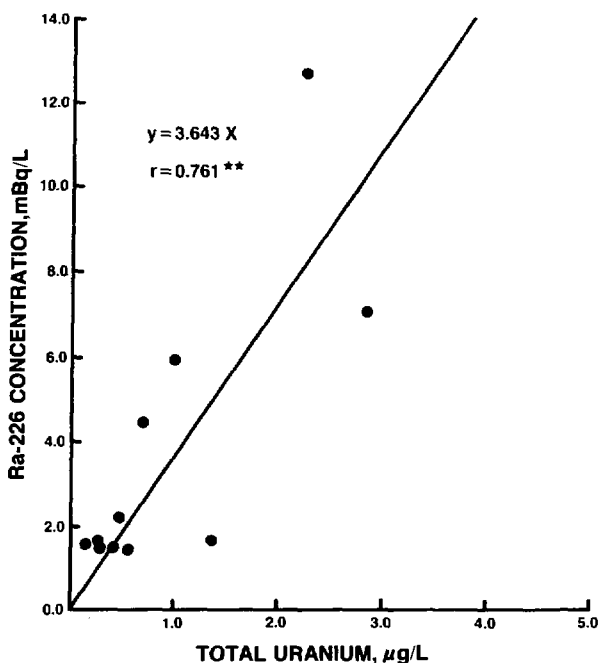


Figure 5. Relationship of total uranium and Ra-226 concentrations in surface waters when plotted through the origin (11 sampling stations; ** indicate $P < 0.01$).

CONCLUSIONS

The levels of radionuclides monitored in selected surface waters are generally low. No time trends were discernible over the monitoring period. Except for total U, at certain locations, all other radionuclide levels were much below the guideline target concentrations for drinking water. Radiological dose commitment from all surface

waters was below the objective dose of $10 \mu\text{Sv}$ (1 mrem), as stipulated in the Great Lakes Water Quality Agreement of 1978. Thus, it may be concluded that the present radionuclide concentrations in the surface waters pose no health hazard at this time.

Limited sediment data suggest that the two nuclear facilities—NPD and Chalk River Nuclear Laboratories—along the Ottawa River have increased the levels of radioactivity in the sediments downstream. More such data are needed from other locations.

Total U and Ra-226 activities suggest that the levels of these radionuclides are related in most surface waters. From their activity ratios, it was concluded that a disequilibrium between the two radionuclides exists, and that total U is much more mobile than Ra-226.

RECOMMENDATIONS FOR FUTURE MONITORING

The available radiological data relating to Canadian surface waters are sparse. The National Radionuclide Monitoring Program has provided a systematic approach to obtaining such data.

Objectives

The objectives of this report were to summarize the available radiological data, identify water quality trends, and assess the environmental significance of the results. The results presented in this report suggest that the program objectives have essentially been achieved.

The upper few millimetres of the sediments are quite reactive (Hart and McKee, 1985; Lerman, 1979; Tracy and Prantl, 1983); the radionuclides can be released into, or removed from, the water column above it. Biota (fish) serves as a foodchain pathway and can also be used as a monitoring tool under certain circumstances.

It is now recommended that the program should be expanded to include associated parameters such as sediments and biota (fish) in order to attain an overall picture of the aquatic ecosystem.

Sampling Locations

At present, up to 13 locations have been monitored for radiological water quality across six provinces and the Northwest Territories. In contrast, only two locations were monitored for sediment quality in Ontario during 1982. As mentioned earlier, most locations were selected because of suspected adverse impacts of uranium/coal mineralization, and/or nuclear fuel cycle operations.

Extensive uranium mineralizations exist in British Columbia, northern Saskatchewan and the Northwest Territories, which are not monitored adequately at present. Particularly in British Columbia, some of the waters surveyed in the areas of Summerland and Nelson have shown concentrations of uranium exceeding the "maximum acceptable concentration" of 20 µg/L. In northern Saskatchewan, very rich uranium deposits exist in the Athabasca Basin. Similarly, besides the Baker Lake area, several other uranium mineralizations exist in the Northwest Territories which are not monitored.

It is, therefore, recommended that the following new stations be added to the NRMP list:

- (i) British Columbia:
 - (a) Kettle River at Midway.
 - (b) Okanagan River at Penticton.
 - (c) Okanagan River below Oliver.
 - (d) Thompson River at Spences Bridge.
- (ii) Northwest Territories:
 - (a) Slave River at Fitzgerald (Alberta).
 - (b) Thelon River below outlet of Schultz Lake.
 - (c) Anigaq River below Audra Lake.
- (iii) Saskatchewan and Manitoba:
 - (a) Beaver River above Beauval.
 - (b) Wollaston Lake at Fond du Lac River and at Cochrane River.
Note: Data from these two stations will be obtained from the Province of Saskatchewan.
 - (c) Sunshine Creek — to be finalized after discussions with Parks Canada.
- (iv) Quebec:
 - (a) Two stations: one above and one below the Nuclear Power Plant at Gentilly.
 - (b) Lac David (southeast of Maniwaki).

Radionuclides and Laboratory Methodology

Radionuclides being analyzed at present are total U, Ra-226, Cs-137, Sb-125 and tritium. Health and Welfare Canada has guidelines for total U, Ra-226, I-131, Cs-137, Sr-90 and tritium; thus I-131 and Sr-90 have been omitted from our list.

Iodine-131 is contributed to surface waters mainly from atmospheric fallout due to nuclear weapons testing and nuclear power generation. This radionuclide has a short half-life of only eight days; hence I-131 poses no long-term hazard. In contrast, Sr-90 is one of the long-lived, health significant, radionuclides in both fallout and effluent discharges from nuclear reactors; its half-life is 28 years.

The Th-232 decay chain radionuclides have only been measured occasionally (Durham and Joshi, 1979b; Great Lakes Water Quality Board, 1983). Recently, it has been increasingly recognized that in spite of low concentrations in waters and sediments, this radionuclide and its daughter products can pose a significant health hazard. In a recent report to Environment Canada, Beak Consultants recommended that Th-232 and its daughter products be included in routine water quality monitoring programs (Hart and McKee, 1985).

Thus, it is recommended that Sr-90 be added to the list of radionuclides being analyzed presently, and that Sb-125 be deleted from the list because of its very low concentrations found in the water to date. It is further recommended that Th-232 and some other thorium isotopes (e.g. Th-230 and Th-228) be added to the list. Inclusion of gross α and β would also be desirable as a screening technique for natural and man-made radionuclides, respectively.

The Radionuclides Section of the National Water Research Institute, which analyzes all of the samples, is keeping up-to-date with the new methodology being developed for analyses. At present, methodologies exist for the analysis of recommended radionuclides, but additional instruments and personnel would be required to carry out the analyses.

Other Radionuclide Databases

Surface water quality data from the Ontario Ministry of the Environment (especially on the Serpent River system), Saskatchewan Department of the Environment, and Saskatchewan Research Council are available. It is recommended that these data be assessed, and pertinent data included in the Water Quality Branch database, NAQUADAT.

Special Problem Areas

Presently, the following areas are experiencing radiological water quality problems:

- (i) Port Radium (Echo Bay)/Rayrock, N.W.T.: These two areas have inactive uranium tailings sites which originated from uranium mining and milling activities in the late 1950s. In 1981, Déné Indians claimed that enough radioactivity had leaked into the waterways such that the foodchain uptake of radionuclides had affected the health of the families living in the area. Health and Welfare Canada has investigated this problem, and preliminary results indicate that Sherman Lake levels for Ra-226 do exceed the "target concentration" for this radionuclide. Sherman Lake eventually discharges into the Marian River System.

- (ii) **Serpent River, Ontario:** The Serpent River system drains the Elliot Lake district where extensive uranium mining and milling activities have been carried out since the early 1950s. The Serpent River discharges through Serpent Harbour into Lake Huron. Several studies have revealed that the levels of radium and other radionuclides in Serpent River water are elevated much above the background levels but are showing a downward trend.

Officials of Health and Welfare Canada, and of the Ontario Ministry of the Environment have expressed interest in Water Quality Branch involvement in the above-mentioned site-specific radiological water quality problems. It is, therefore, recommended that the Water Quality Branch participate in a joint venture with these two agencies.

- (iii) **U.S. nuclear water repository sites near the Canadian border:** The U.S. Department of Energy has announced several sites for consideration to build a nuclear waste repository. Four of these sites in Minnesota are in the watershed of the Red River, and only one site in Maine is in the watershed of the St. Croix River.

It would be desirable to collect background information on these rivers. Already, there is a monitoring station in the Red River, and it is recommended that a station be established in the St. Croix River.

Multi-media Sampling

Sediments and biota (fish) are the other two important components of the aquatic ecosystems. It is, therefore, recommended that these parameters be included in this monitoring program at some selected locations.

Frequency of Sampling

At present, monthly water samples are collected and composited annually. A representative sample is then sent to NWRI for analysis for most sampling locations. To study temporal variations, some samples were analyzed quarterly instead of annually. The results suggest acceptable levels of variability.

Thus, it is recommended that the present scheme for annual composite analyses be followed as before. If an annual composite sample shows anomalous levels of radionuclides, then the samples from that location should be analyzed more frequently. Alternatively, such sampling locations may be sampled more intensively.

Non-radiological Parameters

Radionuclide concentrations in waters are known to be related to some other physico-chemical parameters. It is, therefore, recommended that parameters such as pH, specific conductance and total dissolved solids be measured as well.

Sample Collection Methodology

The present methodology followed for sample collection seems adequate; hence no changes are recommended.

Field Sampling Quality Assurance

A manual relating to field sampling quality assurance is in preparation. It is recommended that any pertinent changes ensuing from this compilation be conveyed to the field staff responsible for sampling.

Laboratory Techniques

Laboratory procedures identified in Chapter 2 are being followed rigorously for quality assurance. Thus the samples collected under the program are analyzed at NWRI using state-of-the-art equipment and techniques (Durham and Joshi, 1979b; Joshi, 1985; Joshi and Durham, 1976). Over the previous three years, emphasis has been placed on the development of reliable low-energy gamma-ray spectrometric techniques for direct (non-destructive) measurement of various radionuclides. At present, these techniques are undergoing rigorous evaluation using recently procured environmental radioactivity standard samples. Such in-house evaluation and participation in various inter-comparison programs are an integral part of all data collected at NWRI. It is therefore recommended that these activities be maintained at the present level.

Interpretive Reporting

Radionuclide data are usually available in the middle of every calendar year for samples collected in the previous year. It is recommended that anomalies be reported annually and that a summary report, comparing the results with the results obtained in the previous years and the literature values, be prepared every three years.

Review of Program

It is recommended that the entire program be reviewed every three years.

Estimate of Resources

Presently, the water samples are collected by regional Water Quality Branch personnel and analyzed by the NWRI Radionuclides Section. As noted earlier, the latter function is an important and expensive aspect of the program. After the samples have been received at NWRI, it requires about 0.25 PY to record, prepare samples, analyze and then report the results to Water Quality Branch headquarters. In addition, it is estimated that chemicals, standard solutions, certified reference materials and instrument maintenance-associated expenditures cost about \$5000 annually. Instrument procurement/replacement costs are not included in these estimates.

If the recommendations outlined in this report are followed, then the water samples alone are likely to double the present number. In addition, if some sediment and fish samples are also included, then the estimated costs would amount to 0.6 to 0.8 PY and an O&M of \$20 000 to \$25 000 annually.

Proposed Implementation

It is recommended that once the decisions are taken, the proposed changes be implemented in the beginning of fiscal year 1986/87.

ACKNOWLEDGMENTS

The National Radionuclides Monitoring Program has been in operation for four years (1981 to 1984). All of the water samples were collected by the regional personnel of the Water Quality Branch; the authors would like to thank them very much for their assistance. S.P. Thompson of the National Water Research Institute is thanked for her assistance in the analytical work, initially carried out under the supervision of R.W. Durham. The helpful suggestions and provision of materials by C. Barraud, P. Vasudev and R. Krauel of Environment Canada, and by D.P. Meyerhof, B.L. Tracy and E. Veska of Health and Welfare Canada are gratefully acknowledged.

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Appendix

Table A-1. Radionuclide Concentration in Surface Waters, 1981

Sampling location*	Radionuclide concentration†				
	Ra-226 (mBq/L)	Cs-137 (mBq/L)	Sb-125 (mBq/L)	Tritium (Bq/L)	Total U (µg/L)
Annapolis River, N.S.	1.85±1.11	-	-	-	0.43±0.06
McAskill Brook, N.S.	1.48±0.37	-	-	3.88±3.22	0.29±0.04
Moose Creek, N.B.	1.48±0.37	-	-	5.81±1.74	1.60±0.20
Petitcodiac River, N.B.	6.66±0.74	-	-	7.33±2.37	1.50±0.20
Niagara River at Niagara-on-the-Lake, Ont.	3.33±0.37	-	-	-	0.39±0.06
St. Lawrence River at Wolfe Island, Ont.	2.22±0.37	-	-	-	0.43±0.06
Saskatchewan River at Man.-Sask. Boundary	7.40±0.74	-	-	9.92±3.22	1.00±0.20
Souris River near Coulter, Man.	2.96±0.37	-	-	15.73±1.74	3.60±0.50
Poplar River East, Sask.	6.66±0.74	-	-	6.48±3.22	3.40±0.50
Baker Lake, N.W.T.	1.11±0.37	-	-	17.46±1.74	0.13±0.02
Mean	3.52	-	-	9.52	1.28

* Sampling locations are indicated in Figure 3.

† The errors reported with each measurement are the statistical errors of the individual measurements including those in the background.

Table A-2. Radionuclide Concentration in Surface Waters, 1982

Sampling location*	Radionuclide concentration†				
	Ra-226 (mBq/L)	Cs-137 (mBq/L)	Sb-125 (mBq/L)	Tritium (Bq/L)	Total U (µg/L)
Ranger Bight, Lab.	4.44±0.37	13.69±1.85	<5.55	8.62±2.15	0.73
Annapolis River, N.S.	1.11±0.37	<1.85	<5.55	4.96±2.15	0.41
McAskill Brook, N.S.	1.48±0.37	6.66±1.48	8.88±4.07	<2.18	0.25
Moose Creek, N.B.	1.11±0.37	<1.85	8.88±4.44	6.03±2.15	1.43
Petitcodiac River, N.B.	18.50±0.74	4.44±1.85	<3.70	8.18±2.15	0.33
Saskatchewan River at Man.-Sask. Boundary	6.29±1.11	<2.98	<8.35	-	1.05
Souris River near Coulter, Man.	3.33±0.37	<1.11	5.92±4.07	4.96±1.92	0.93
Poplar River East, Sask.	7.03±0.74	<2.53	5.52±4.73	-	3.16
Baker Lake, N.W.T.	1.48±0.37	2.96±1.11	5.18±4.07	14.84±1.92	0.10
Niagara River at Niagara-on-the-Lake, Ont.	2.34±1.02	0.92±0.38	<1.81	8.68±2.48	0.50±0.11
St. Lawrence River at Wolfe Island, Ont.	1.31±0.38	1.76±0.81	<2.40	10.44±1.00	0.98±0.98
Mean	4.40	-	-	-	0.90

* Sampling locations are indicated in Figure 3.

† The errors reported with each measurement are the statistical errors of the individual measurements including those in the background.

Table A-3. Radionuclide Concentration in Surface Waters, 1983

Sampling location*	Radionuclide concentration†				
	Ra-226 (mBq/L)	Cs-137 (mBq/L)	Sb-125 (mBq/L)	Tritium (Bq/L)	Total U (µg/L)
Ranger Bight, Lab.	5.55±0.74	<3.48	<7.78	4.29±1.74	0.61±0.09
Annapolis River, N.S.	1.48±0.37	3.97±2.55	8.98±7.40	7.07±1.74	0.50±0.02
McAskill Brook, N.S.	2.22±0.74	<2.56	22.24±8.47	5.51±1.74	0.34±0.05
Moose Creek, N.B.	2.59±0.37	<3.39	<8.42	9.47±1.81	5.77±0.86
Petitcodiac River, N.B.	23.68±0.74	<2.57	<6.93	10.06±1.81	4.84±0.73
Souris River near Coulter, Man.	1.48±0.37	<2.86	<6.93	5.99±1.74	2.14±0.32
Red River, Man.	6.66±0.37	8.57±3.03	<7.52	6.33±1.74	6.90±1.03
Baker Lake, N.W.T.	2.59±0.37	<4.29	<7.39	9.10±1.81	0.29±0.04
Niagara River at Niagara-on-the-Lake, Ont.	1.36±0.67	0.47±0.26	<1.16	8.32±2.00	0.58±0.05
Niagara River at Fort Erie, Ont. ‡	1.85±0.11	1.00±0.32	1.33±0.91	3.66±2.37	0.16±0.08
St. Lawrence River at Wolfe Island, Ont.	1.09±0.31	1.29±0.21	<1.50	7.26±1.53	0.45±0.04
Mean	4.60	-	-	7.01	2.05

* Sampling locations are indicated in Figure 3.

† The errors reported with each measurement are the statistical errors of the individual measurements including those in the background.

‡ The data are only for the 4th Quarter.

Table A-4. Radionuclide Concentration in Surface Waters, 1984

Sampling location*	Radionuclide concentration†				
	Ra-226 (mBq/L)	Cs-137 (mBq/L)	Sb-125 (mBq/L)	Tritium (Bq/L)	Total U (µg/L)
Ranger Bight, Lab.	3.33±0.37	<2.58	<5.35	4.94±2.96	0.76±0.11
Annapolis River, N.S.	1.48±0.37	<2.28	<5.32	<2.96	0.39±0.06
McAskill Brook, N.S.	0.74±0.37	<2.30	<8.56	<2.96	0.32±0.05
Moose Creek, N.B.	1.48±0.37	<2.60	<6.99	5.93±2.96	1.09±0.16
Petitcodiac River, N.B.	1.85±0.37	<3.08	7.17±5.74	5.93±2.96	2.29±0.34
Souris River near Coulter, Man.	0.74±0.37	4.46±2.48	<4.62	3.87±2.96	2.88±0.43
Red River, Man.	1.11±0.37	2.44±2.19	<6.36	<2.96	2.58±0.39
Saskatchewan River at Man.-Sask. Boundary	4.07±0.37	<4.63	<7.84	<2.96	0.91±0.14
Poplar River East, Sask.	7.40±0.74	<3.50	<7.24	<2.96	1.97±0.30
Baker Lake, N.W.T.	1.11±0.74	5.47±5.01	<10.19	7.58±3.05	0.14±0.02
Niagara River at Niagara-on-the-Lake, Ont.	1.73±0.56	<0.73	<1.28	7.00±0.87	0.45±0.03
Niagara River at Fort Erie, Ont.	1.48±0.30	<0.50	<0.77	6.58±1.58	0.40±0.07
St. Lawrence River at Wolfe Island, Ont.	1.11±0.07	<1.01	<1.17	7.97±1.53	0.43±0.05
Mean	2.13	-	-	-	1.12

*Sampling locations are indicated in Figure 3.

†The errors reported with each measurement are the statistical errors of the individual measurements including those in the background.

Table A-5. Whole Body Radiological Dose Commitment Factors Used in This Report

Radionuclide	Annual limit of intake (Bq)	Committed effective dose equivalent for 1 mBq/L concentration in water (µSv)
Radium-226	1.7×10^5	2.36×10^{-1}
Cesium-137	3.7×10^6	1.09×10^{-2}
Antimony-125	7.6×10^7	5.28×10^{-4}
Tritium	2.9×10^9	1.38×10^{-5}
Total uranium	6.4×10^5	6.27×10^{-2}

Note: Annual limit of intake values were taken from the IJC 1983 Report on Great Lakes Water Quality – Appendix on Radioactivity, except for natural (total) uranium for which the value given by Johnson and Dunford (1983) was used.

Table A-6. Whole Body Radiological Dose Commitment from Total Uranium in Surface Water When Used as a Source of Drinking Water

Sampling location*	Total U concentration		Committed effective dose equivalent (µSv)
	µg/L	mBq/L†	
1. Ranger Bight, Lab.	0.70	17.90	1.12
2. Annapolis River, N.S.	0.43	10.99	0.69
3. McAskill Brook, N.S.	0.30	7.67	0.48
4. Moose Creek, N.B.	1.37	35.03	2.20
5. Petitcodiac River, N.B.	2.24	57.27	3.59
6. Niagara River at Niagara-on-the-Lake, Ont.	0.48	12.27	0.77
7. Niagara River at Fort Erie, Ont.	0.28	7.16	0.45
8. St. Lawrence River at Wolfe Island, Ont.	0.57	14.57	0.91
9. Souris River near Coulter, Man.	2.39	61.10	3.83
10. Red River, Man.	4.74	121.19	7.60
11. Saskatchewan River at Man.-Sask. Boundary	0.99	25.31	1.59
12. Poplar River East, Sask.	2.84	72.61	4.55
13. Baker Lake, N.W.T.	0.16	4.09	0.26

*Sampling locations are indicated in Figure 3.

†An equilibrium among the uranium isotopes is assumed: 1 µg of total U = 25.567 mBq (0.691 pCi).

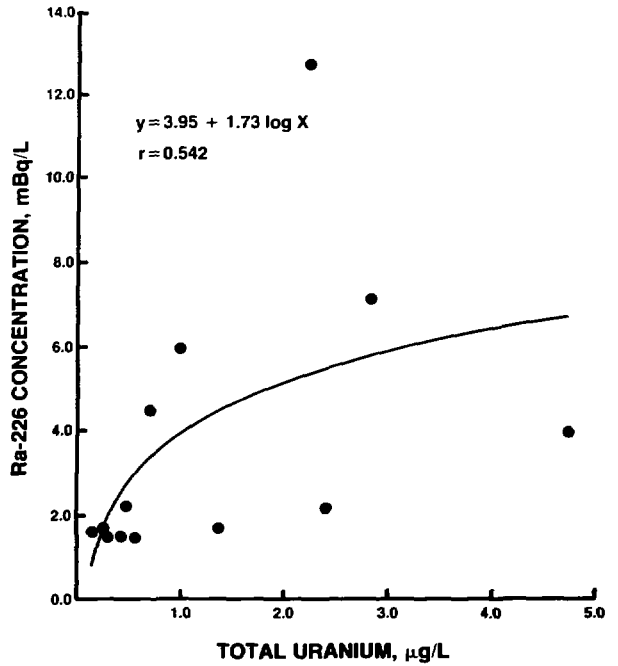


Figure A-1. Relationship of total uranium and Ra-226 concentrations in surface waters (13 sampling stations).

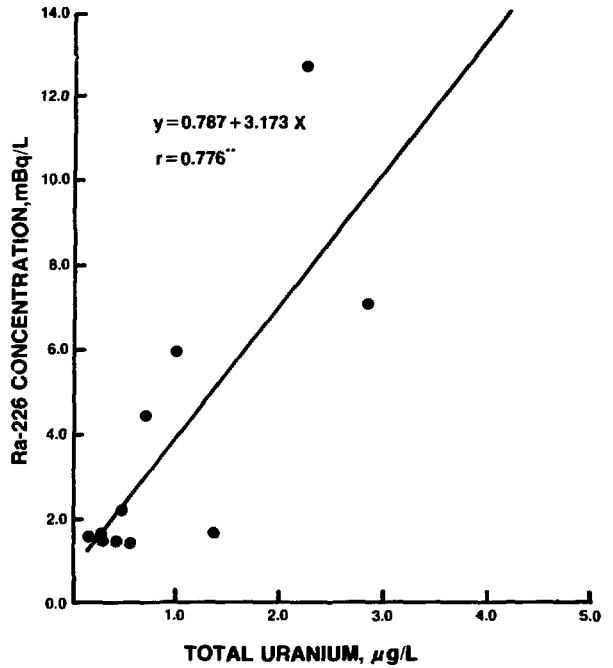


Figure A-2. Relationship of total uranium and Ra-226 concentrations in surface waters (11 sampling stations).