Determination of Radioactivity Levels in the Great Lakes: Final Report

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DSS Contract Serial No. : OSS-00020

DETERMINATION OF RADIOACTIVITY LEVELS IN THE GREAT LAKES

FINAL REPORT

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June 7,1978

Final Report Accepted

Dr. R.W.Durham
Scientific Authority

June 19,1978

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1. INTRODUCTION

This report describes the laboratory work done pursuant to the titled contract.The samples for radiochemical analyses,carried out in the NWRI Radiochemistry Laboratories, were provided by the Scientific Authority of the project, Dr.R.W.Durham,who coordinated their collection and delivery. All the salient features of this report have earlier been communicated to the Scientific Authority in the form of regular monthly progress reports.Some of the methods used and the results obtained during the course of these studies have been reported earlier and,therefore,their presentaion in this report is limited.

2. METHODS

2.1 Water samples for ^{137}Cs, ^{125}Sb, ^{90}Sr, ^{3}H,and ^{226}Ra

The radionuclides ^{137}Cs and ^{125}Sb were determined by gamma-ray spectral analysis. The CCIW techniques,described by Durham (1974),were employed for these analyses except that no stable caesium carrier was added to the samples.

For the determination of the ^{90}Sr content, the sample containing previously added Sr carrier (50-100 mg) was diluted, $Sr(CO_3)_2$ precipitated,and then dissolved in minimum HNO_3.Ba (10 mg) carrier was then added and Ca removed by cyclic 72% HNO_3 precipitations and alcohol-ether separations.The nitrates were then dissolved in water and Ba removed as $BaCrO_4$. $Sr(CO_3)_2$was precipitated, redissolved in 1M HNO_3,and ^{90}Y removed by $Fe(OH)_3$ scavenging. $Sr(CO_3)_2$ was reprecipitated,set aside for atleast 15 days,dissolved in 1M HCl,and Y carrier (10 mg) added.Y was separated and purified from Sr through a cycle of hydroxide precipitations and HNO_3 dissolutions.Finally,$Y(C_2O_4)_3.9H_2O$ was precipitated and beta-counted on a low background proportional counter.The ^{90}Sr content was calculated from the ^{90}Y net count data ,corrected for yields and efficiency.During the conse of these analyses it was observed that Ca is not quantitatively removed by 72% HNO_3 precipitations and alcohol-ether separations.Therefore,the final yields were obtained through AA analysis technique in lieu of the standard gravimetric method.

For the determination of ^{3}H, an 8-ml aliquot of the water sample was thoroughly mixed with 15 ml of scintillation cocktail 'PCS' (Amersham Corporation) in a polyethylene scintillation vial until a proper counting gel was formed. The vial was then placed in the counting chamber and allowed to reach equilibrium overnight before counting. Sample counting data was then corrected for background, and ^{3}H content obtained by using counter efficiency data.

^{226}Ra in the 4-l water samples was determined by ^{222}Rn emanation method, described elsewhere (Durham and Joshi, 1977).

2.2 Fish samples for ^{137}Cs and ^{226}Ra

A 250-300 g (wet wt.) fish sample was sliced and kept soaked at room temperature overnight in about 50 ml of conc.HNO_3. The volume was then reduced to about 25 ml and the wet ashing completed by using 35% H_2O_2 and conc.HNO_3. The sample was then taken to dryness and final volume made to 40 ml using dil.HNO_3. The sample was first gamma counted for ^{137}Cs and then diluted to 130 ml for the determination of ^{226}Ra by emanation method mentioned earlier.

2.3 Sediment samples for ^{137}Cs and ^{60}Co

A known amount (usually 40g) of the dried, groud sample was gamma counted on the Ge(Li) detector, previously standardized for these radionuclides. The amounts of these radionuclides were determined using net count in their respective photopeaks, detector efficiency, and pertinent nuclear data.

3. RESULTS

3.1 Water samples

3.1.a The 1976 and 1977 levels of ^{137}Cs, ^{125}Sb, and ^{90}Sr are presented in Tables I and II. The errors given are statistical errors derived from sample and background counting data.

3.1.b During the course of an investigation of Lake Ontario water quality near Port Granby radioactive waste management site, 124 water samples were analyzed for ^{226}Ra. These results have already been communicated in

the form of a CCIW Unpublished Report (Durham and Joshi,1977) and are,therefore,not included in this report.Briefly,these results indicated that ^{226}Ra is leaching from the said waste management site and is reaching Lake Ontario waters.

3.1.c One sample,provided by IAEA under its International Intercomparison Programme,was analyzed for ^{3}H,^{90}Sr,and ^{137}Cs.These results are given in Table III.

3.2 Fish samples

Ten Lake Ontario fish(Rainbow Trout),collected in April 1976 from the mouth of Ganaraska River,were analyzed for ^{137}Cs and ^{226}Ra.These results are presented in Table IV.

3.3 Sediment samples

Two sediment samples,provided by IAEA under its International Intercomparison Programme,were analyzed for ^{60}Co and ^{137}Cs.The results are presented in Table V.

Table I. Levels of ^{137}Cs and ^{125}Sb in Great Lakes Waters, 1976

Lake	Sample Station			Depth in Metres	Date	Level (pCi m^{-3})	
	Northern Latitude	Western Longitude	No.			^{137}Cs	^{125}Sb
Superior	47°02'20"	85°05'54"	3	1	10/6	54±7	<10
				75	10/6	55±6	<10
				147	10/6	49±6	<10
	47°50'12"	87°27'00"	12	1	12/6	49±5	38±12
				110	12/6	57±5	28±11
				220	12/6	63±5	19±9
	47°12'24"	89°40'00"	21	1	16/6	52±5	<10
				100	16/6	47±5	34±12
				200	16/6	47±5	36±11
Huron	45°15'00"	82°53'00"	11C	1	20/6	35±5	41±12
	45°43'12"	83°17'30"	11D	1	20/6	23±3	31±8
	43°30' 00"	82°04'12"	10D	1	21/6	17±3	38±10
Ontario	43°15'39"	79°11'39"	13	1	4/12	18±3	9±8
	43°22'54"	77°17'54"	40	1	5/12	23±4	23±11
	43°43'54"	77°01'06"	69	1	6/12	14±3	38±8
St.Clair	43°27'30"	82°46'48"	-	1	22/6	18±4	33±9

Table II. Levels of ^{90}Sr, ^{125}Sb, and ^{137}Cs in Great Lakes Waters, 1977

Lake	Sample Station			Depth in Metres	Date	Level (pCi/l)		
	Northern	Western	No.			^{90}Sr	^{125}Sb	^{137}Cs
Huron	43°43'00"	81°57'00"	-	1	8/8/77	0.84±0.03	0.060±0.015	0.038±0.00
Erie	42°34'30"	79°36'36"	14	1	1/9/77	0.69±0.02	0.041±0.014	0.014±0.00
				55	1/9/77	-	0.028±0.012	0.017±0.00
	42°09'00"	81°18'30"	76	1	1/9/77	0.95±0.02	0.028±0.010	0.030±0.00
				26	1/9/77	0.78±0.03	0.060±0.014	0.022±0.00
Ontario	43°25'02"	79°24'03"	13	1	18/8/77	0.84±0.02	< 0.025	<0.010
				103	18/8/77	0.95±0.03	0.030±0.011	0.019±0.00
	43°35'40"	78°00'50"	40	1	18/8/77	1.14±0.02	0.038±0.013	0.026±0.00
				178	18/8/77	0.90±0.02	0.040±0.013	0.039±0.00
	43°36'24"	76°42'42"	69	1	17/8/77	0.86±0.02	0.054±0.012	0.029±0.00
				185	17/8/77	0.89±0.02	0.052±0.012	0.017±0.00

Table III. Results for the analysis of IAEA water sample

Radio-nuclide	unit	Net results of individual determinations						Random error resulting from counting statistics (%).	Maximum uncertainty due to assessable systematic error (%)
		1	2	3	4	5	6		
^{3}H	nCi/l	2.01	1.99	1.95	1.90	1.97	1.97	5	3
^{90}Sr	pCi/l	3.01	2.92	3.46	–	–	–	8	6
^{137}Cs	pCi/l	1.00	1.10	1.04	–	–	–	15	6

Table IV. Levels of ^{137}Cs and ^{226}Ra in Lake Ontario fish (Rainbow Trout) from mouth of Ganaraska River

Collection Date	Mass of Whole fish(Kg)*	Sex	Concentration, pCi/Kg (wet wt.)	
			^{137}Cs	^{226}Ra
April 17, 1976	10.0	-	76±3	3.8±0.3
	2.77	M	53±3	17.0±0.6
	2.50	M	51±3	2.6±0.3
	3.00	M	69±3	1.4±0.2
	2.45	M	85±5	60.2±1.2
	3.40	F	68±3	< 0.2
	3.18	F	62±3	2.5±0.2
	3.81	F	62±5	44.7±0.9
	3.00	F	65±4	0.4±0.1
	0.36	F	44±6	71.5±2.5

*analyses performed on posterior sections

Table V. Results of radionuclide analysis on IAEA sediment samples

Sample	Radio-nuclide	Dry weight (g)	Concentration (pCi/g) on reference date (1/1/1977)	Estimated error (pCi/g)
SD-B-2	^{60}Co	40	260	13
	^{137}Cs	40	72.1	2.5
SD-B-3	^{60}Co	40	0.22	0.05
	^{137}Cs	40	154	5

4. The development of a method for the determination of isotopic thorium in solids and solutions from uranium mining and milling operations.

During the course of an EPS/NWRI collaborative project on the determination of several naturally-occuring radionuclides in various solutions and solids originating from uranium mining and milling operations, a need arose for the unambiguous determination of ^{228}Th, ^{230}Th, and ^{232}Th in these matrices. Several approaches toward the development of a reliable method for this purpose were examined and a procedure was eventually carved out of the results of these experiments. A detailed account of these investigations (to be published in a learned journal at a subsequent date) has already been submitted to the Scientific Authority of the project.

Briefly, the method consists in the determination of ^{228}Th via monitoring of the 239-keV gamma-emission of its daughter ^{212}Pb. To determine the concentrations of ^{230}Th and ^{232}Th, the thoriums are selectively extracted using high molecular weight amines and electrodeposited on stainless-steel backing prior to the recording of their alpha spectra. From an analysis of alpha spectral data for ^{228}Th, ^{230}Th, and ^{232}Th in conjunction with results from the gamma spectrometric determination of ^{228}Th, the concentrations of individual thorium isotopes are easily obtained.

This method was then used to analyze a number of leachate, sediment, tailings, and miscellaneous samples. The analyses on leachate samples were performed by Mr. J.A. FitzGerald, whereas the author of this report analyzed the remaining samples the results of which are given in Table VI.

Table VI. Results of isotopic thorium analyses .

Sample	Concentration (pCi/g)		
	^{228}Th	^{230}Th	^{232}Th
038A Tailings	6.5	3.2	1.6
038A Sediment	162	3790	332
038B Sediment	28	607	56
038C Tailings	34	402	34
035 Sludge	0.45	-	-
035 Buckwheat	<0.01	-	-
Grass 10 loam (Al high)	<0.01	-	-
Grass 16 loam (Fe high)	<0.01	-	-

5. ACKNOWLEDGEMENTS

The author is indebted to Dr.R.W.Durham for advice and useful discussions during the course of this work. Grateful acknowledgement is also made to Mr.R.J.Goble for detector calibrations and tritium analyses.

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