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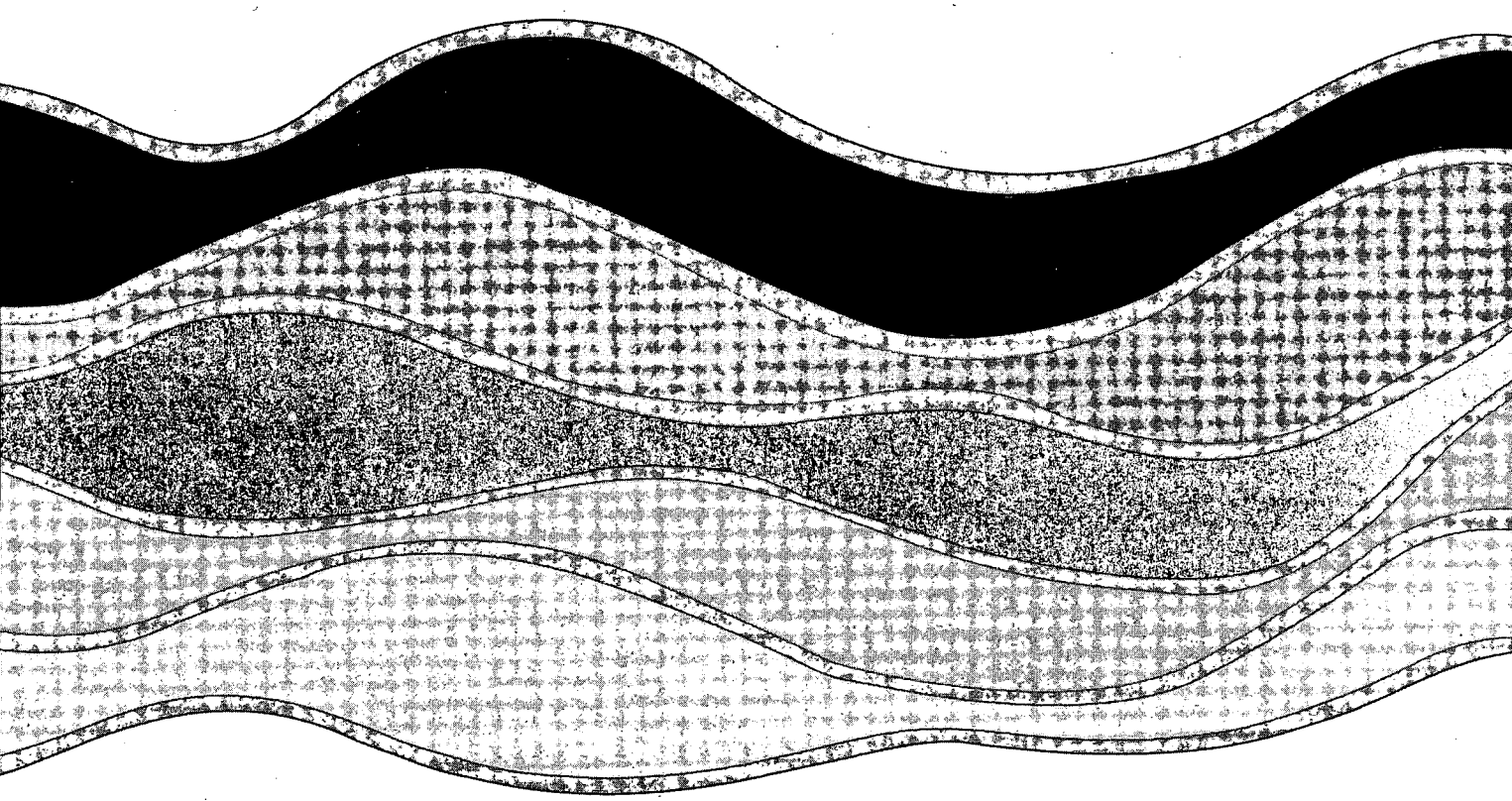

SOURCES AND BEHAVIOR OF ANTHROPOGENIC RADIONUCLIDES IN THE OTTAWA RIVER WATERS

S.R. Joshi and R.C. McRea

NWRI Contribution No. 91-09

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Management Perspective

The present study was initiated by NWRI, and carried out jointly with WQB-Ontario Region, to assess the levels and transport behaviour of radionuclides released from the Chalk River Nuclear Laboratories (CRNL) to the Ottawa River. The concentrations of several gamma-ray emitting radionuclides were measured in raw water, centrifuged water, drinking water and suspended solid samples collected from the Ottawa River near Ottawa between 1984-86. In addition, water filtration plant floc samples from a nearby location were also analyzed. The data on radionuclide levels suggest the allowable release limits have not been approached.

Two of the major nuclear fission products detected, ^{90}Sr and ^{137}Cs, were also released during the atmospheric testing of nuclear weapons during 1958-63. Each of the two radionuclides has a half-life of about 30 years which means that nearly half of the activity released still resides on land. Our analysis of the historical CRNL release data and available monitoring data indicates that most of the ^{90}Sr in the river derives from its fallout inventory in the watershed rather than from the CRNL facilities. On the other hand, both nuclear fallout and CRNL-derived ^{137}Cs contribute equally to the prevailing levels of this radionuclide in the river.

The data also show that while some fraction of ^{137}Cs will be retained on the floc during filtration of the receiving waters, hard ß-emitting ^{90}Sr will pass through to the drinking water. (By comparison, ^{137}Cs is a soft ß-emitter.) At present, the radiation dose to the public via this route is insignificant and, in fact, is lower than that provided by naturally-occurring ^{226}Ra present in the drinking water.

The presence of fallout ^{90}Sr and ^{137}Cs provides a unique opportunity to evaluate their mean residence times and to evaluate soil erosion rates in the watershed. Thus far, we have worked out some of the mathematical groundwork necessary for this purpose. The residence time information could be of use in studying the behaviour of diffuse source heavy metal inputs to the watershed.

Perspectives de la direction

La présente étude a été amorcée par l'INRE et effectuée en collaboration avec le Conseil de la qualité de l'eau-région de l'Ontario; l'étude a pour but d'évaluer la concentration et le transport des radionucléides rejetés dans la rivière des Outaouais par les Laboratoires nucléaires de Chalk River (LNCR). Les concentrations de plusieurs radionucléides émetteurs de rayons gamma ont été mesurées dans l'eau brute, l'eau centrifugée, l'eau potable et des échantillons de matières en suspension prélevés dans la rivière des Outaouais près d'Ottawa entre 1984 et 1986. Des échantillons de floc d'usine de filtration d'eau prélevés dans les environs ont également été analysés. Les résultats concernant la concentration des radionucléides indiquent que les limites admissibles de rejet n'ont pas été approchées.

Deux des principaux produits de fission nucléaire détectés, le ^{90}Sr et le ^{137}Cs, ont également été dégagés lors des essais nucléaires effectués dans l'atmosphère entre 1958 et 1963. Les deux radionucléides ont une demi-vie d'environ 30 ans, ce qui signifie que presque la moitié de l'activité dégagée est encore présente sur le sol. Notre analyse des données historiques de rejets des LNCR et les données de surveillance dont nous disposons actuellement indiquent que la plupart du ^{90}Sr présent dans la rivière provient plutôt des retombées dans le bassin hydrographique que des rejets des installations des LNCR. Par contre, dans le cas du ^{137}Cs, les retombées nucléaires et les installations des LNCR sont également responsables des concentrations actuelles de ce radionucléide dans la rivière.

Les données montrent également que, si une partie du ^{137}Cs, qui est un émetteur de rayons ß mous, est retenue sur le floc au cours de la filtration des eaux réceptrices, le ^{90}Sr, qui est un émetteur de rayons ß durs, passe outre et se retrouve dans l'eau potable. À l'heure actuelle, la dose de rayonnement que reçoit la population par cette voie est insignifiante et est en fait inférieure à la dose provenant du ^{226}Ra présent naturellement dans l'eau potable.

La présence de ^{90}Sr et de ^{137}Cs due aux retombées nous fournit l'occasion unique de calculer leur temps de séjour et d'évaluer le taux d'érosion du sol dans le bassin hydrographique. Nous avons jusqu'à maintenant effectué une partie des calculs mathématiques à cette fin. Les données sur le temps de séjour pourraient nous être utile dans l'étude des apports de métaux lourds de sources mal identifiées dans le bassin hydrographique.

Abstract

Multiphase radionuclide measurements on the Ottawa River waters are reported for the period October 1984 to March 1986. Numerous radionuclides are present in detectable amounts in the raw, drinking and centrifuged waters as well as in the suspended sediment and water filtration plant floc samples. The sediment/water partitioning behavior of these radionuclides is also reported. It is observed that the prevailing low particle flux allows rapid migration of radionuclides through the system. It is also found that most of the ^{90}Sr in the river derives from its fallout inventory in the watershed rather than from the Chalk River Nuclear Laboratories (CRNL). On the other hand, both the fallout and CRNL-derived ^{137}Cs appear to contribute equally to the prevailing levels of this radionuclide in the river. The data also suggest that the prevailing levels of the naturally-occurring ^{226}Ra contribute more than those of ^{90}Sr insofar as the radiological quality of the drinking water is concerned. Many of the radionuclides are removed to the floc during filtration of the receiving waters. Their removal efficiencies, however, cannot be precisely defined from the currently available measurements.

Résumé

On présente les concentrations de radionucléides mesurées à plusieurs époques dans les eaux de la rivière des Outaouais pour la période d'octobre 1984 à mars 1986. Un grand nombre de radionucléides sont présents en concentrations décelables dans l'eau brute, l'eau potable et l'eau centrifugée, ainsi que dans des échantillons de sédiments en suspension et de floc d'usine de filtration de l'eau. On présente également le mode de partage de ces radionucléides entre les sédiments et l'eau. On a observé que le faible flux actuel des particules permet une migration rapide des radionucléides dans le système. On a également observé que la plupart du ^{90}Sr présent dans la rivière provient plutôt des retombées radioactives dans le bassin hydrographiques que des Laboratoires nucléaires de Chalk River (LNCR). Par ailleurs, il semble que les retombées et les LNCR soient également responsables des concentrations actuelles de ^{137}Cs dans la rivière. Les données indiquent également que les concentrations actuelles de ^{226}Ra d'origine naturelle ont plus d'effet que celles de ^{90}Sr sur la qualité radiologique de l'eau potable. Un grand nombre de radionucléides sont retenus dans le floc lors de la filtration des eaux réceptrices. Toutefois, les données dont nous disposons actuellement ne nous permettent pas de définir avec précision l'efficacité de rétention.

1. Introduction

With a drainage basin of approximately 146,000 km^2, the Ottawa River is the largest tributary of the St. Lawrence River. Certain segments of the 1150-km long river, particularly those below Ottawa, are known to be polluted with agricultural, industrial and municipal wastes (Environment Canada - Environnement Quebec - Environment Ontario, 1985). The river has also received fallout from the atmospheric testing of nuclear weapons and radioactive effluents from two separate nuclear installations located about 200 km upstream from Ottawa. Once released, the nuclear facility-generated radioactivity is transported downstream (Roy *et al.*, 1990) to major population centres.

Although Ottawa River waters have been regularly monitored for radioactivity (Cooper, 1985; Health and Welfare Canada, 1973-86; Ontario Hydro, 1976-86), no attempt has been made to clearly discern the relative contribution of the fallout and nuclear facility-derived radionuclides in the system. The present communication gives an account of multiphase measurements performed on the river waters during 1984-86 to assess the relative significance of these two major sources of artificially-produced radionuclides in the basin. Results from the water/sediment partitioning studies of the radionuclides are also presented. And finally, we assess the removal of radionuclides on aluminium hydroxide floc during treatment of raw water for drinking purposes. Results from companion measurements on the partitioning and transport of two major naturally-occurring radionuclides, ^{210}Pb and ^{226}Ra, in this river system have been reported earlier (Joshi *et al.*, 1991).

2. Site Description and Methods

The Ottawa River basin is shown in Figure 1. Discharge is regulated through numerous dams and other control structures. Over the course of the river, the mean annual discharge increases from 450 $m^3 s^{-1}$ at the head of Lake Temiskaming to about 2060 $m^3 s^{-1}$ near its confluence with the St. Lawrence River (Peter Yee, Water Planning & Management Branch, Inland Waters Directorate, Burlington, personal communication, 1987). About 9.9% of the drainage basin is comprised of waterbodies. The major portion of the river drains the Canadian Shield, and therefore a relatively high content of natural radionuclides can be expected. The south-eastern portion of the basin is characterized by sedimentary limestone bedrock. The average annual precipitation in the watershed is about 870 mm, about 50% of which is released in the form of runoff (OWRC - QWB, 1971).

A multiphase approach was adopted for radionuclide monitoring of the Ottawa River at Lemieux Island (Ottawa). The waters in this area are well-mixed due to a combination of several sets of rapids immediately upstream; the site also permits easy midstream sampling. The river is typically 3 to 4 m deep in the vicinity of the sampling site and has a limestone bed virtually free of bottom sediments. McCrea and Fischer (1986) have described the technical aspects of the monitoring station.

Raw water, centrifuged water, and suspended sediment samples were collected on a weekly or biweekly basis and subsequently composited on a quarterly basis

from October 1984 to March 1986. Phase separation was achieved with a Westfalia continuous centrifuge operating at 9500 g. The method provides a near-quantitative recovery of suspended solids. The water samples were collected in acid-washed polyethylene bottles and then preserved with nitric acid. Drinking water and floc samples were obtained from the Lemieux Island water treatment plant. The plant uses aluminium sulphate and sodium silicate treatment for flocculation and coagulation purposes.

The 30 to 50 L water samples were reduced to a counting volume of 40 mL with the addition of stable Sr as carrier and yield monitor for ^{90}Sr. The water and freeze-dried suspended sediment and floc samples (5 to 10 g) were analyzed by high-resolution gamma-ray spectrometry using hyperpure Ge detectors in planar and coaxial configurations. Each sample was counted for 2.5×10^5 s or longer. The characteristics of the detectors and details of the quality assurance procedures have been given earlier (Joshi, 1985, 1987, 1989). The system background was substantially reduced by enclosing the detector in shielding cage constructed out of 1 to 2 mm thick layers of stainless steel and metallic Cu, Cd, Al and Sn. This cage in turn is surrounded by 10-cm thick pre-World War II lead. The multilayer shielding dramatically reduces the background, particularly in the low-energy region. With this arrangement the 'background' contribution of radionuclides of interest is essentially negligible as determined by frequent 5×10^5s long counts. However, care should be exercised in interpreting the gamma-ray spectra since use of cadmium lining gives rise to two small but detectable emissions at 558 and 651 keV while reducing the flux of cosmic ray neutrons which interact with In and Ge (Roy *et al.*, 1989). The radionuclide

concentrations were computed as

$$\frac{(T - B) \pm \sqrt{T + B}}{F} ,$$

where T = total counts in the peak channels,

B = background counts (taken as average of equal number of channels on either side of the peak), and

F = factor used to convert count rate to disintegration rate and includes count time, detection efficiency, sample size and gamma-ray emission probability.

If A denotes the net counts in the peak due to radionuclide under consideration, then $A = T - B$ and $\sigma_A = \sqrt{T + B} = \sqrt{A + 2B}$. When low levels of radioactivity are involved, $A \rightarrow 0$ and $\sigma_A \rightarrow \sqrt{2B}$. Therefore, the counting error quoted later in the text pertains to "greater than $\sqrt{2B}$" since, in actual practice T is much higher than B so that $\sigma_A >> \sqrt{2B}$. It should be noted that B refers only to the Compton continuum under the photopeak since, as noted earlier, the special passive shielding used results in negligible 'background' contribution due to radionuclide of interest. In the absence of such measures, 'background' radionuclide contribution to the photopeak should also be subtracted to obtain the net counts.

The analytical procedure for the determination of ^{90}Sr has been described by Durham and Joshi (1984). All data reported are corrected for decay to the mid-point of the sample collection period.

3. Sources of Anthropogenic Radionuclides to the Ottawa River

3.1 Delivery of fallout radionuclides

Anthropogenic radionuclides have been introduced into the Ottawa River basin since about 1945. The number and magnitude of the atmospheric tests began increasing in the early 1950's when measurements of fallout delivery also commenced. The amount of this activity increased significantly in the late 1950's and, after a brief pause, peaked during 1962-63 prior to a limited test ban treaty coming into force. Since then few atmospheric tests have been conducted in the northern hemisphere by the People's Republic of China. Over the last decade or so, the fresh fallout deposition of ^{90}Sr and/or ^{137}Cs has more or less equalled the decay of their radioactivity stored in the environment. Our estimates of the delivery of these two fallout radionuclides to a mid-Ottawa River basin location are given in Table I. These estimates were derived from regression analysis (Joshi, 1991) of data on the deposition of fallout radioactivity in North America (Larsen, 1985); the levels during 1984-86 were assumed to be similar to those in 1983.

3.2 Releases from nuclear facilities

The Chalk River Nuclear Laboratories (CRNL) of the Atomic Energy of Canada Limited are located on the Ontario bank of the Ottawa River, about 190 km upstream from Ottawa (Figure 1). Operating since 1945, the CRNL comprises four nuclear reactors and diverse research, isotope production, and radioactive waste management facilities. The Nuclear Power Demonstration (NPD) reactor, Canada's first nuclear power reactor, is located at Rolphton, about 25 km upstream from CRNL. Commissioned in 1962, the NPD generating station operated until 1987. Both CRNL and NPD have introduced artificially-produced radionuclides into the Ottawa River.

It has been estimated that the CRNL released a total of 27 TBq of gross ($\beta+\gamma$) activity via its liquid effluents during 1972-80 (NRCC, 1983). During the same period, the NPD generating station's liquid effluents contained 20 GBq of gross ($\beta+\gamma$) activity, or about 0.07% of the CRNL releases. Thus, for the purpose of the present study, the contribution of NPD releases to the Ottawa River system may be deemed negligible.

Five liquid effluent streams discharge into the Ottawa River at CRNL (Cooper, 1985); of these only the process sewer can be considered as an important source of radioactivity. The process sewer discharges at a rate of 1.6 $m^3\ s^{-1}$ and contains several fission and activation products from reactor operations as well as radioactive effluent from decontamination and waste treatment activities. Table II presents the release history of ^{90}Sr and ^{137}Cs from

the process sewer to the Ottawa River for the period of 1969 to 1985. These estimates were derived by summing the daily release data given in the quarterly progress reports released by the CRNL (AECL, 1969-86).

4. Results and Discussion

4.1 Water/sediment partitioning of radionuclides

Results of the raw water, drinking water, centrifuged water, and suspended sediment analyses are given in Table III. In addition to the listed radionuclides, ^{59}Fe, ^{65}Zn, ^{95}Zr-^{95}Nb, ^{103}Ru, ^{106}Ru, ^{110m}Ag, ^{125}Sb, ^{141}Ce, ^{155}Eu, and ^{241}Am were also detected on occasion. All the fission and activation products listed in Table III, or otherwise detected at irregular intervals, were released from the CRNL (AECL, 1984-86) during the study period. In some cases, the non-detection of certain radionuclides which were regularly discharged to the river may be due to their decay during long time intervals between sample collection and analysis. Non-detection may also result from extremely low concentrations obtainable due to dilution in the river waters. The water/sediment partitioning and transport characteristics of ^{210}Pb and ^{226}Ra have been discussed earlier (Joshi *et al.*, 1991).

The affinity of radionuclides for suspended sediments is usually inferred

from the values of field distribution coefficient, K_d (mL g^{-1}), calculated as the ratio between radioactivity in the suspended sediment to that in the dissolved phase (centrifuged water). The values of K_d obtained for representative radionuclides in the Ottawa River system are given in Table IV. These values reflect the relative affinities of various radionuclides for the particulate matter in water. The extent of radionuclide transport in the particulate phase will also be determined by the concentration of particles in the system. If f represents the fraction of the radioactivity in the suspended sediment and c the concentraion of suspended sediment (g mL^{-1}), it can be shown that

$$1/f = 1 + 1/ (K_d \, . \, c) \qquad (1)$$

Using equation (1) and the average values of K_d (Table IV) and c (Table V), we calculate that f ranges from about 0.001 for ^{90}Sr to about 0.27 for ^{137}Cs (and ^{210}Pb). This clearly shows that essentially all ^{90}Sr is in the dissolved form. The partitioning of the particle-reactive radionuclides such as ^{60}Co and ^{137}Cs in the dissolved form undoubtedly stems from the low particle concentration in the river.

An insight into the rapidity with which radionuclides migrate through the system may be gained through an examination of the behavior of ^{60}Co. This radionuclide is generally deemed to be a non-fallout activation product though it may also be produced through the $^{60}Ni(n,p)^{60}Co$ reaction during a thermonuclear explosion in which the required 14-MeV neutrons are known to be generated and the precursor ^{60}Ni may be provided by the device casing. Indeed Roy *et al.*(1981)

have detected such ^{60}Co following an atmospheric nuclear test by the People's Republic of China. The amount of activity detected however was very small, less than 1% of that due to ^{137}Cs. Considering the relatively short half-life of this radionuclide, the general lack of atmospheric testing of nuclear weapons during the study period, and its activity levels vis-a-vis those of ^{137}Cs (Table III), we may realistically assume that all ^{60}Co in the river arises from the CRNL.

From the data given in Tables III and V, we estimate (Table VI) that nearly all (~99%) of the ^{60}Co supplied by the CRNL site is rapidly exported through our sampling location. It is possible that part of this ^{60}Co is derived from bottom sediments (i.e., relatively 'old' ^{60}Co) which are known (Baweja *et al.*, 1987) to contain CRNL-delivered ^{60}Co. However, since about 74% of the ^{60}Co activity migrates in the dissolved phase (Table VI), it is likely that only a small fraction of the total activity can be attributed to bed load or resuspended bottom sediments. Therefore, it seems logical to conclude that nearly all CRNL-delivered ^{60}Co transports through our sampling site without significant time lag. Roy *et al.*(1990) have recently estimated that the CRNL-delivered radionuclides, including ^{60}Co, transit from Chalk River to Ottawa in 22 to 42 d. The present results appear to be in line with these measurements.

4.2 Fallout *versus* CRNL-delivered ^{90}Sr and ^{137}Cs in the Ottawa River

Two of the more consistently detected radionuclides, ^{90}Sr and ^{137}Cs, are also released in significant amounts during the atmospheric testing of nuclear

weapons. Although the direct flux of this radioactivity has substantially decreased in the past two decades or so (Table II), their relatively long half-lives ensure their persistence in the area soils from where they may be released to the river. These two radionuclides are expected to display different geochemical interactions in the environment as ^{90}Sr is chemically conservative while ^{137}Cs exhibits strong association with various minerals. Their disparate interactions are reflected in the K_d values given in Table IV.

The concentrations of CRNL-derived radionuclides in the Chalk River area of the Ottawa River may be estimated from the radionuclide discharge data given in Table II and the water flow charateristics of the Ottawa River at the nearby Des Joachims gauging station (Environment Canada, 1987). These estimates are shown in Figure 2. In addition to the CRNL-produced radionuclides, the Ottawa River waters also contain fallout radioactivity directly impinging on the river water surface. As the river has low particle concentrations, much of the ^{137}Cs and almost all the ^{90}Sr thus received by the river will likely be quickly transported. If we assume an average width of 1 km for the river upto the Chalk River area, then the annual concentrations of these two radionuclides in the river can be readily estimated from the fallout data (Table I) and the annual water discharge in the area (Environment Canada, 1987). These estimates are also shown in Figure 2.

Since the early 1960s, Health and Welfare Canada has monitored the Ottawa River waters for both ^{90}Sr and ^{137}Cs at five locations between Rolphton and Ottawa. Meyerhof (1984) has reported the radionuclide concentrations in the samples

collected until 1983. Although not exactly specified, the raw water sampling site appears to be located downstream from Chalk River. These data are also shown in Figure 2; the values for 1984-86 were obtained from the published (HWC, 1984-86) monitoring data.

The results given in Figure 2 clearly show that the two radionuclides behave differently in the watershed. First and foremost, it is observed that while all measured ^{137}Cs may be accounted for by that falling directly on the river surface and by that arising from the CRNL facilities, the same cannot be said about ^{90}Sr which largely seems to derive from the watershed. A time lag of about two yr between the estimated direct input and measured concentration peaks for the latter radionuclide is indicative of rapid removal from the watershed. Brown (1961) has earlier reported a fallout ^{3}H-based mean residence time of 3.7 yr for water in this drainage basin. The enhanced mobility of ^{90}Sr, as indicated by its low K_d value (Table IV), is quite evident from the data in Figure 2 from which it can be seen that although fallout ^{137}Cs input exceeds that of ^{90}Sr (by a factor of 1.6) it is ^{90}Sr which is largely leaching from the watershed.

These results also imply that the area soils efficiently sequester fallout ^{137}Cs and its removal from the drainage basin is directly linked to the slow erosion of soils. Thus ^{137}Cs has a much longer watershed residence time when compared with ^{90}Sr. An evaluation of this mean residence time is beyond the scope of the present communication, but it should be very similar to that for ^{210}Pb (Joshi *et al.*,1991) which K_d value similar to that for ^{137}Cs.

The results given in Figure 2 also show that since 1970-71, the measured ^{137}Cs concentrations have been at about the levels expected from the combined direct fallout and CRNL inputs. Undoubtedly, some ^{137}Cs does derive from the drainage basin but as yet we are unable to precisely estimate this contribution which should be very small. The presently available results suggest that the current levels of ^{137}Cs in this river cannot be exclusively attributed to a single dominant source. On the other hand, the results for ^{90}Sr clearly show that the fallout activity stored in the drainage basin is still the prime contributor to the continuously declining levels of this radionuclide in the river.

4.3 Removal of radionuclides during water filtration

As part of the water filtration process, aluminium hydroxide is used to clarify the raw water. The resulting floc removes a significant proportion of the dissolved organic matter and other waterborne contaminants including radionuclides. Analysis of the floc material has been used to estimate radionuclide concentrations in the entering waters (Roy *et al.*, 1979; Durham and Joshi, 1981). This technique undoubtedly provides much higher sensitivity for those radionuclides which are efficiently retained by the floc, though even in such cases quantification of the results is often difficult. This approach was used by Roy *et al.*(1990) in the evaluation of the radionuclide levels in the Ottawa and the St. Lawrence Rivers.

The results of our measurements on the floc samples, given in Table VII,

show that essentially the same radionuclides as are present in the water samples (Table III) are detected in the floc samples. Their retention on the floc was estimated using the relationship

$$\text{Percent retained} = (1 - C_{dw}/C_{rw}) \times 100 \quad , \qquad (2)$$

where C_{dw} and C_{rw} denote the average radionuclide concentrations in the drinking water and raw water, respectively. Table VIII summarizes the results obtained using concentrations in water averaged over the first three quarters of 1985 which correspond to the floc sample measurements. This approach, although logical in design, may also lead to ambiguous results as is evident from Table VIII where we find that the levels of several radionuclides in drinking water exceed those in the raw water thus implying zero retention on the floc. In each case, however, we do detect these radionuclides in the floc. This could be attributable to the sampling protocol adopted or to the analytical errors or to a combination of both.

The relative contribution of each possible source of error cannot be precisely delineated at the moment, but this situation is quite common in partitioning studies where the levels in the dissolved and the particulate phases often do not add up to those in the raw water as is also the case with the data in Table III. In the present study, many of our measurements on water samples approach the detection limit of the instrument. This can obviously lead to situations where a radionuclide though present at extremely low levels is reported as 'not detected' with the implicit suggestion that it might be

altogether absent. This is best illustrated using the example of ^{238}U which is essentially reported as 'not detected' (that is, it is below the detection limit) in both the raw water and the drinking water samples (Table III). A simplistic interpretation would tend to assume that the radionuclide is altogether absent in the system whereas both the suspended sediment (Table III) and the floc (Table VII) sample measurements attest to the contrary. The easy detection of this radionuclide in the latter two matrices undoubtedly arises from the fact that samples of both correspond to much higher water sample volumes than is the case with direct measurements on the water samples. Thus we believe caution must be exercised in deriving such information solely on the basis of measurements on water samples.

Alternatively, the removal of a radionuclide by the floc may be qualitatively assessed by considering its concentrations in the suspended sediment and the floc where the sensitivity issue does not hamper interpretation. The basis for this approach stems from the studies reported by Turcotte *et al.* (1984) which show that the concentration of ^{137}Cs in the floc is proportional to the concentration of the particulate matter in the entering waters. Lupien and Grondin (1984) also report a similar correlation for the naturally-occurring ^{228}Ra. Following these observations, the concentration of the radionuclide *x* in floc, $C_{x,floc}$ (Bq g^{-1}), may be given as

$$C_{x,floc} = k_x \times C_{x,ss} \times C_{ss,w} \times 1/C_{floc,w} \quad , \qquad (3)$$

where k_x is the proportionality factor which may indicate the influence of

processes other than the simple removal of the fraction associated with the suspended sediment in the entering waters, $C_{x,ss}$ is the concentration of the radionuclide in the suspended sediment (Bq g^{-1}), $C_{ss,w}$ is the concentration of suspended sediment in the entering waters (g mL^{-1}), and $C_{floc,w}$ the concentration of flocculating particles in the water (g mL^{-1}). Unfortunately, the values of $C_{floc,w}$ are not available to us to derive an estimate of k_x. The parameters $C_{floc,w}$ and $C_{ss,w}$ may, however, be eliminated by writing an equation analogous to equation (3) for the radionuclide *y*

$$C_{y,floc} = k_y \times C_{y,ss} \times C_{ss,w} \times 1/C_{floc,w} \qquad (4)$$

From equations (3) and (4), we obtain

$$k_y/k_x = C_{y,floc} \times C_{x,ss} / C_{x,floc} \times C_{y,ss} \qquad (5)$$

If we designate ^{137}Cs as the reference radionuclide *x* , any significant deviation from unity in the value of k_y/k_x should imply that factors other than the simple entrapment of suspended solids by the flocculating particles are responsible for the retention of a given radionuclide *y* on the floc. Results of our calculations using data (Tables III and VII) pertaining to the first three quarters of 1985 are given in Table IX. These calculations clearly suggest that like ^{137}Cs, the radionuclides ^{60}Co, ^{90}Sr and ^{144}Ce are largely removed to the floc in association with suspended sediments, while other processes also play a significant role in the retention of the naturally-occurring radionuclides, particularly ^{228}Th and ^{238}U. It should be noted that the enrichment of a

radionuclide in floc vis-a-vis suspended sediment may also be ascribed to the possible contamination of flocculating agents with the radionuclide under consideration in the first place. From our experience with the detection of extremely low levels of radioactivity in several materials used in the construction of shielding for gamma-ray detectors and in laboratory reagents, such a possibility cannot be ruled out. The currently available data, however, are inadequate in assessing this scenario.

4.4 Estimated distribution of ^{90}Sr and ^{137}Cs in the Ottawa River fish

Besides water and sediments, some of the radioactivity in the river is undoubtedly incorporated in the plants and fish. We are not aware of any data on radioactivity in the aquatic plants, but the available (HWC, 1973-76; Ontario Hydro, 1976-86) measurements on various species of fish when assessed in the light of their relative summer abundance (ORPG, 1979) suggest that an extremely small fraction (of the order of 10^{-4} to 10^{-3}%) of ^{137}Cs or ^{90}Sr activities transported through the system is carried by the fish. This, however, does not imply that only insignificant additional radiation risk is associated with the consumption of the river fish as the same data also show that the fish/water radionuclide concentration factors are about 300 and 5600 for ^{90}Sr (bone) and ^{137}Cs (flesh), respectively.

5. Conclusions

The data collected in the present study show that both fallout and nuclear facility-derived radionuclides are present in the waters and suspended sediments of the Ottawa River. Our analysis of these data indicates that even the particle-reactive radionuclides such as ^{60}Co and ^{137}Cs are quickly transported through the system. We also conclude that most of the ^{90}Sr currently present in the river is released from the fallout ^{90}Sr inventory in the soils. The same, however, cannot be said about ^{137}Cs where both fallout and CRNL-derived fractions appear to make equal contributions to the total.

Measurements on the water filtration plant floc samples show that much of the ^{90}Sr and ^{137}Cs present in the raw water eventually reaches the drinking water supply. The retention of several of the radionuclides by the floc shows peculiar behavior which cannot be adequately explained using the limited measurements afforded by the present investigation. The data, however, are sufficient to infer that the low particle concentration prevailing in the river results only in the partial retention of some particle-reactive anthropogenic radionuclides which may provide additional radiation dose to the public in the event of a large unplanned release from the CRNL facilities. At the moment these radionuclides contribute very little to the low radiation dose received by the area residents drinking the river water. In fact, in terms of the radiological quality of the drinking water (HWC, 1987), our data suggest that the prevailing levels of naturally-occurring ^{226}Ra in the river waters are more significant than those of

^{90}Sr. And finally, our analysis of existing data shows that very small fractions of the ^{90}Sr and ^{137}Cs activities in the river are incorporated in the fish. The fish/water radionuclide concentration factors, however, do not suggest that only insignificant additional risk may be associated with the consumption of the river fish.

Acknowledgements

We thank the NWRI Engineering Services Section and the staff of the Regional Municipality of Ottawa-Carleton for invaluable help at various stages of the study. J. Fischer, H. DaGama and the staff of the Lemieux Island water filtration plant assisted in sample collection. S.P. Thompson and J.A. FitzGerald are thanked for performing the laboratory analyses. Numerous discussions with Professor J.-C. Roy (Laval University) on analytical measurements and the floc sampling technique are gratefully acknowledged.

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TABLE I

Estimated annual delivery of fallout ^{90}Sr and ^{137}Cs to a mid-Ottawa River basin location[a] (46° 30' N)

Year	Delivery (MBq km^{-2}) ^{90}Sr	^{137}Cs
1954	66.0	105.6
1955	85.4	136.6
1956	105.8	169.3
1957	106.2	169.9
1958	147.3	235.6
1959	207.3	331.8
1960	37.5	60.6
1961	58.1	92.9
1962	294.8	471.7
1963	568.9	910.3
1964	379.9	606.5
1965	132.2	211.5
1966	57.9	60.7
1967	39.3	62.9
1968	31.5	50.4
1969	34.2	54.8
1970	35.3	56.6
1971	33.8	54.1
1972	18.0	28.7
1973	9.9	15.9
1974	22.3	35.6
1975	16.3	26.1
1976	3.6	5.7
1977	19.4	31.0
1978	20.5	32.9
1979	6.9	11.0
1980	3.4	5.4
1981	8.6	13.8
1982	1.0	1.6
1983	1.0	1.6
1984	1.0	1.6
1985	1.0	1.6
1986 (Jan.-March)[b]	0.3	0.4

[a]A $^{137}Cs/^{90}Sr$ activity ratio of 1.6 was used in processing the raw data.

[b]Deposition rate during 1984-86 is assumed to be the same as in 1983.

TABLE II

Estimated annual releases of ^{90}Sr and ^{137}Cs from CRNL to the Ottawa River[a]

Year	Activity (GBq) ^{90}Sr	^{137}Cs
1969	179.0	108.0
1970	70.4	138.0
1971	55.5	104.0
1972	33.8	41.8
1973	46.1	50.3
1974	54.2	54.2
1975	67.3	70.9
1976	78.3	112.0
1977	54.9	147.0
1978	31.7	54.8
1979	45.7	87.1
1980	24.2	107.0
1981	27.0	109.0
1982	32.0	23.8
1983	29.0	242.0
1984	110.0	102.0
1985	10.1	53.5
1986 (Jan.-March)	3.7	13.0

[a]Computed from the reported (AECL, 1969-86) daily release data.

TABLE III

Levels of radionuclides in water and suspended sediment samples. ND, not detected; NM, not measured.

Sampling period	Activity (mBq L^{-1})			Suspended sediment (mBq g^{-1}dry)
	Raw water	Drinking water	Centrifuged water	
Anthropogenic Radionuclides				
	^{46}Sc (half-life, 83.8 d)			
1984,4Q	ND	ND	2.4±1.3	ND
1985,1Q	ND	ND	ND	ND
2Q	ND	ND	2.7±1.3	ND
3Q	3.3±1.4	ND	1.9±1.3	96±24
4Q	ND	ND	3.8±1.2	1943±58
1986,1Q	ND	ND	ND	NM
	^{54}Mn (half-life, 312 d)			
1984,4Q	ND	ND	ND	57±3
1985,1Q	0.7±0.3	ND	0.6±0.3	63±2
2Q	ND	5.8±0.5	ND	26±2
3Q	ND	ND	0.5±0.3	38±5
4Q	0.8±0.5	ND	0.6±0.4	416±12
1986,1Q	1.4±0.6	ND	ND	NM
	^{60}Co (half-life, 5.3 y)			
1984,4Q	0.8±0.3	0.8±0.3	0.4±0.3	59±2
1985,1Q	0.9±0.3	1.1±0.2	0.8±0.2	107±2
2Q	0.6±0.2	2.0±0.3	0.3±0.2	34±2
3Q	0.5±0.2	0.4±0.2	0.7±0.2	37±4
4Q	ND	ND	0.5±0.3	82±7
1986,1Q	ND	ND	ND	NM
	^{90}Sr (half-life, 29 y)			
1984,4Q	21.5±0.3	18.5±0.3	21.8±0.4	NM
1985,1Q	16.3±0.3	14.6±0.3	16.2±0.3	NM
2Q	12.8±0.3	12.0±0.3	12.9±0.3	NM
3Q	18.7±0.4	16.4±0.3	16.7±0.3	12.2±7.7
4Q	17.2±0.5	18.1±0.6	19.6±0.5	7.4±3.7
1986,1Q	18.4±0.4	17.4±0.4	17.0±0.4	NM

contd./...

^{137}Cs (half-life, 30.1 y)

1984,4Q	4.6±0.3	3.4±0.3	1.7±0.3	582±3
1985,1Q	3.6±0.3	2.1±0.3	2.6±0.2	320±3
2Q	3.0±0.2	1.6±0.2	2.0±0.3	300±3
3Q	4.3±0.3	3.0±0.2	3.8±0.2	414±6
4Q	1.6±0.4	1.2±0.4	2.0±0.3	406±6
1986,1Q	3.5±0.5	0.9±0.4	3.7±0.5	NM

^{144}Ce (half-life, 284 d)

1984,4Q	ND	ND	2.0±1.4	153±9
1985,1Q	ND	ND	1.5±0.8	204±10
2Q	2.7±1.0	ND	ND	31±6
3Q	ND	ND	ND	53±11
4Q	7.8±2.2	ND	4.5±1.9	836±22
1986,1Q	3.2±1.1	ND	3.0±1.9	NM

Naturally-Occurring Radionuclides

^{7}Be (half-life, 53 d)

1984,4Q	76±15	ND	ND	39700±5400
1985,1Q	ND	ND	ND	ND
2Q	ND	ND	97±42	484±223
3Q	ND	ND	120±60	982±398
4Q	ND	ND	ND	ND
1986,1Q	ND	ND	ND	NM

^{210}Pb (half-life, 22.3 y)

1984,4Q	3.4±1.2	ND	2.2±0.3	500±9
1985,1Q	3.3±1.0	ND	3.5±1.2	521±10
2Q	3.8±1.2	2.7±1.2	4.1±1.0	361±9
3Q	3.5±1.2	ND	1.1±0.3	432±16
4Q	ND	ND	5.9±1.7	516±23
1986,1Q	5.0±1.6	ND	3.8±1.3	NM

^{226}Ra (half-life, 1620 y)

1984,4Q	4.7±1.4	3.1±1.2	5.8±1.5	30±3
1985,1Q	ND	2.8±1.2	3.4±1.1	30±3
2Q	3.6±1.0	11.2±1.2	5.9±1.1	29±3
3Q	3.9±1.1	5.1±1.0	2.0±1.0	35±7
4Q	6.1±2.1	8.4±2.1	7.6±1.8	17±9
1986,1Q	7.4±1.4	9.0±1.9	4.1±1.2	NM

contd/...

	^{228}Th (half-life, 1.9 y)			
1984,4Q	1.9±0.5	2.3±0.7	1.9±0.7	68±3
1985,1Q	1.4±0.4	2.0±0.5	1.0±0.5	54±3
2Q	ND	5.0±0.7	1.6±0.5	54±3
3Q	ND	0.9±0.4	0.8±0.4	60±5
4Q	ND	1.2±0.8	ND	66±6
1986,1Q	ND	1.2±0.6	1.0±0.5	NM
	^{238}U (half-life, 4.5 x 10^9 y)			
1984,4Q	ND	2±1	ND	41±6
1985,1Q	ND	ND	ND	23±6
2Q	ND	ND	ND	15±6
3Q	ND	ND	ND	20±9
4Q	3±2	ND	ND	ND
1986,1Q	ND	ND	ND	NM

TABLE IV

Field distribution coefficients of representative radionuclides in the Ottawa River system. NM, not measured; NA, not applicable as no activity was detectable in the water sample.

Sampling period	K_d (mL g^{-1})			
	Anthropogenic Radionuclides			
	^{60}Co	^{90}Sr	^{137}Cs	^{144}Ce
1984, 4Q	$1.5x10^5$	NM	$3.4x10^5$	$7.7x10^4$
1985, 1Q	$1.3x10^5$	NM	$1.2x10^5$	$1.5x10^5$
2Q	$1.1x10^5$	NM	$1.5x10^5$	NA
3Q	$5.3x10^4$	731	$1.1x10^5$	NA
4Q	$1.6x10^5$	379	$2.0x10^5$	$1.9x10^5$
1986, 1Q	NM	NM	NM	NM
	Naturally-Occurring Radionuclides			
	^{7}Be	^{210}Pb	^{226}Ra	^{228}Th
1984, 4Q	NA	$2.3x10^5$	$5.2x10^3$	$3.6x10^4$
1985, 1Q	NA	$1.5x10^5$	$8.8x10^3$	$5.4x10^4$
2Q	$5x10^3$	$8.8x10^4$	$4.9x10^3$	$3.4x10^4$
3Q	$8x10^3$	$3.9x10^5$	$1.8x10^4$	$7.5x10^4$
4Q	NA	$8.7x10^4$	$2.2x10^3$	NA
1986, 1Q	NM	NM	NM	NM

TABLE V

Mean discharge[a] and suspended sediment concentrations in the Ottawa River

Sampling period	Discharge ($m^3\ s^{-1}$)	Total discharge (m^3)	Susp.sed. concn. ($mg\ L^{-1}$)	Total sediment export (kg)
1984, 4Q	1159	9.20×10^9	1.72	1.58×10^7
1985, 1Q	1453	1.13×10^{10}	1.47	1.66×10^7
2Q	2070	1.63×10^{10}	4.39	7.15×10^7
3Q	872	6.95×10^9	1.01	6.98×10^7
4Q	859	6.83×10^9	1.45	9.96×10^6
1986, 1Q	1097	8.51×10^9	1.10	9.93×10^6

[a]Environment Canada (1987)

TABLE VI

Export of CRNL-derived ^{60}Co through the sampling site; NA, not available

Sampling period	Input[a] (GBq)	Output (GBq)		
		Dissolved	Suspended	Total
1984, 4Q	5.2	3.7	0.9	4.6
1985, 1Q	NA	9.0	1.8	10.8
2Q	8.5	4.9	2.4	7.3
3Q	NA	4.9	0.3	5.2
4Q	2.5	3.4	0.8	4.2

[a]AECL (1984-86)

TABLE VII

Levels of radionuclides in the water filtration plant floc samples; ND, not detected

Sampling date	Activity (mBq g^{-1}dry)				
	Anthropogenic Radionuclides				
	^{46}Sc	^{60}Co	^{90}Sr	^{137}Cs	^{144}Ce
February 13,1985	ND	ND	ND	63±3	22±8
March 13, 1985	ND	14±2	4±1	36±2	40±9
April 17,1985	30±18	17±2	4±1	129±3	14±5
May 1, 1985	40±19	9±2	ND	107±2	16±5
June 15, 1985	ND	ND	ND	84±3	17±4
July 9, 1985	ND	5±2	4±2	97±5	ND
September 25,1985	53±7	35±4	4±2	56±19	8±3
	Naturally-Occurring Radionuclides				
	^{7}Be	^{210}Pb	^{226}Ra	^{228}Th	^{238}U
February 13,1985	ND	160±8	15±3	77±3	56±8
March 13, 1985	ND	312±9	15±2	46±11	76±5
April 17, 1985	ND	339±10	18±3	116±3	79±6
May 1, 1985	900±400	305±9	16±2	128±3	53±6
June 15, 1985	900±400	236±8	7±2	108±3	50±6
July 9, 1985	ND	249±18	23±7	87±7	47±6
September 25,1985	850±200	153±6	8±2	83±2	49±5

TABLE VIII

Retention of radionuclides by the water filtration plant floc as given by relationship (2)

Radionuclide	Percent retained
^{60}Co	nil
^{90}Sr	10
^{137}Cs	38
^{144}Ce	~100
^{210}Pb	74
^{226}Ra	nil
^{228}Th	nil

TABLE IX

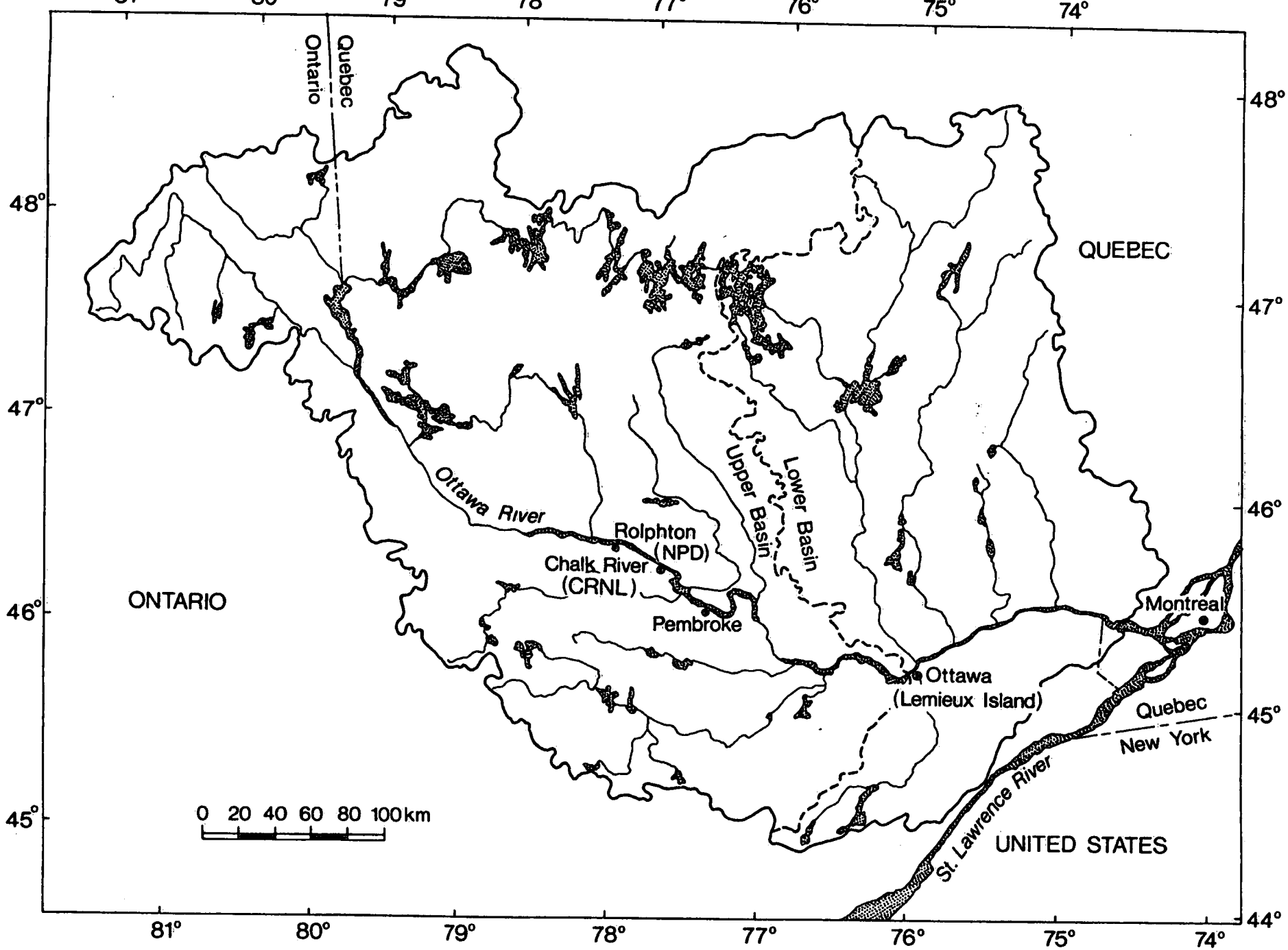
Relative significance of the processes responsible for the retention of radionuclides on floc as indicated by equation (5) using ^{137}Cs as the reference radionuclide(x). If entrapment of the suspended sediment is the sole process involved, the ratio k_y/k_x approaches unity.

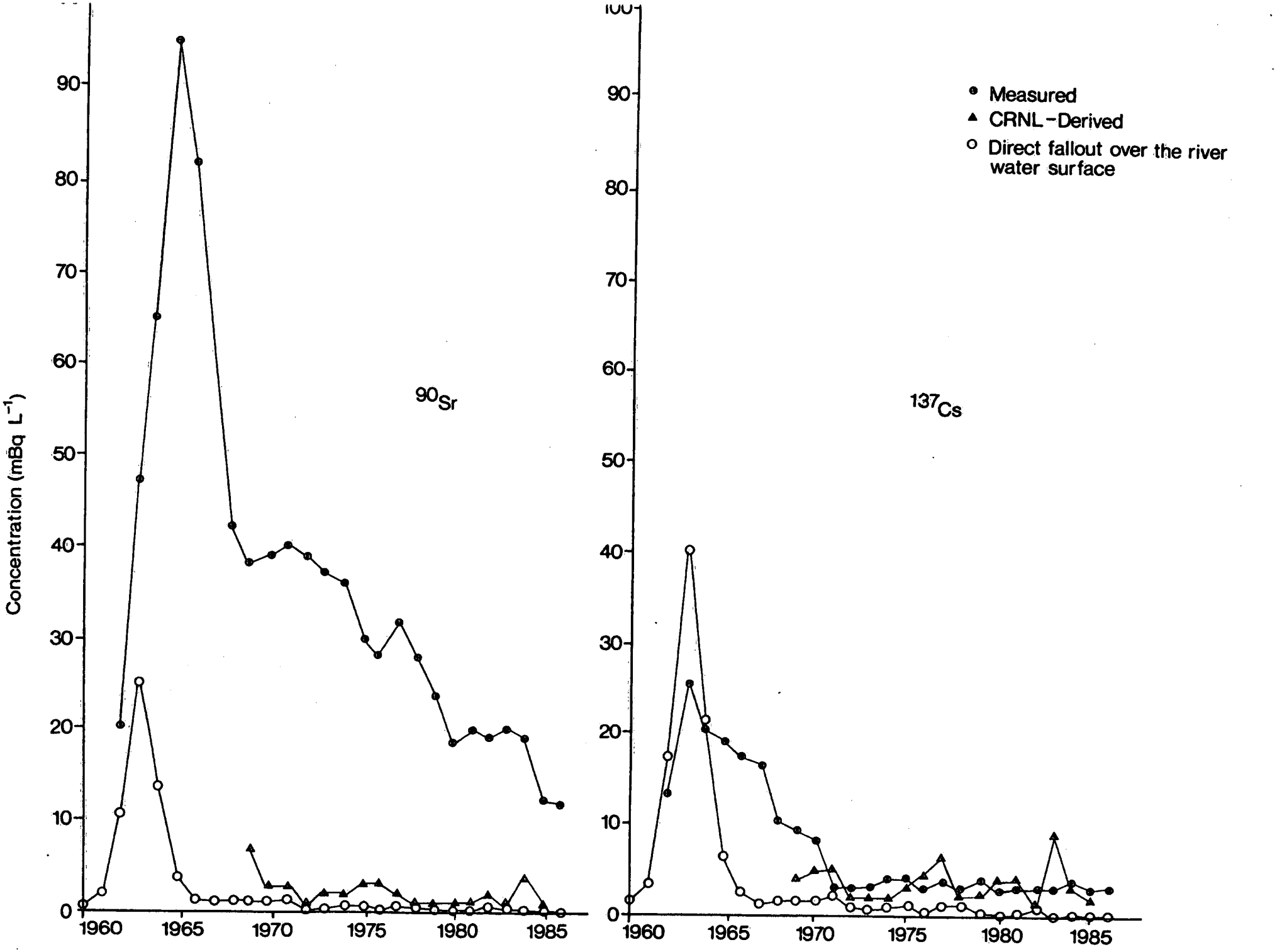
Radionuclide	k_y/k_x
^{60}Co	0.9
^{90}Sr	1.0
^{137}Cs	-
^{144}Ce	0.8
^{210}Pb	2.6
^{226}Ra	2.2
^{228}Th	7.5
^{238}U	14.1

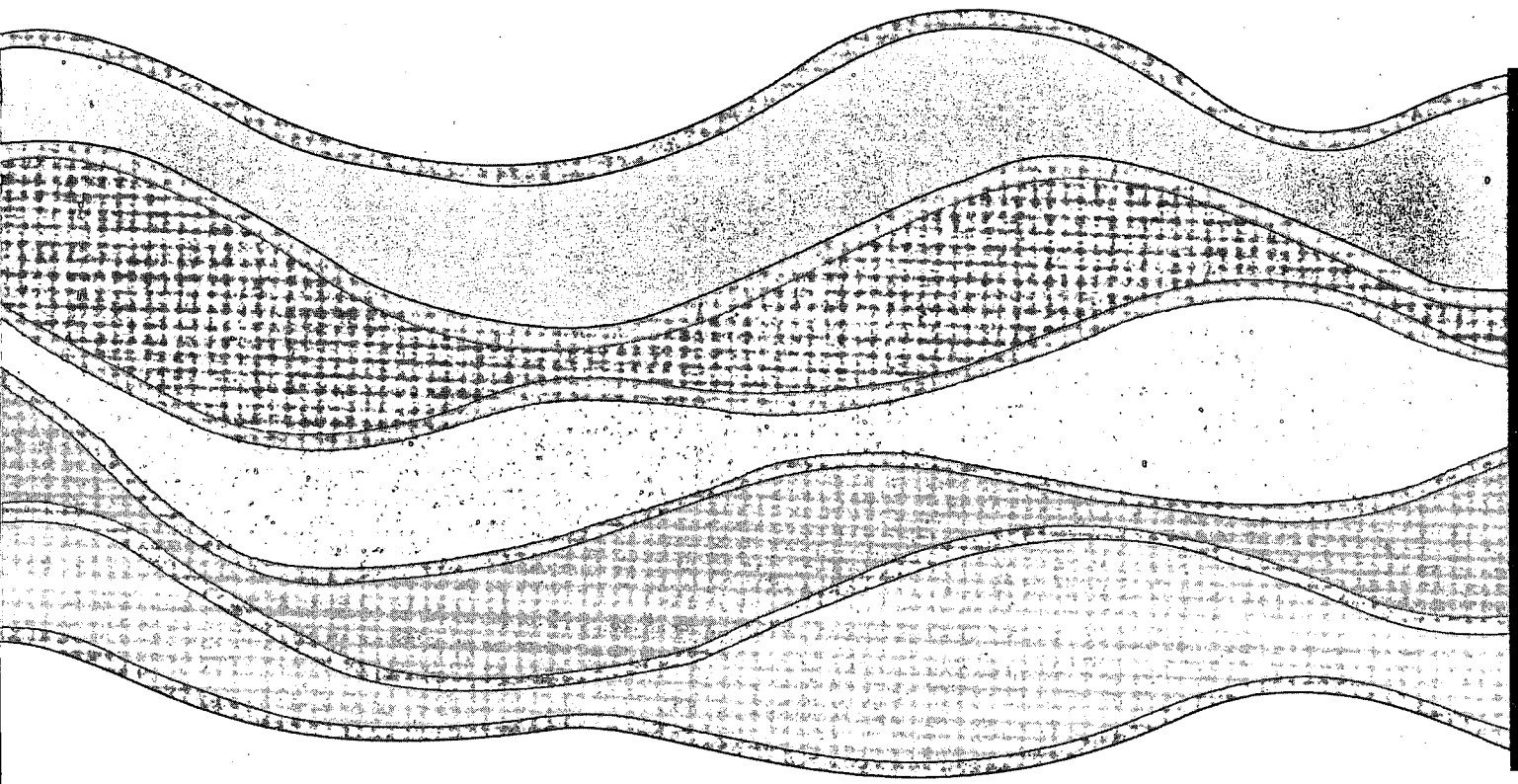
CAPTIONS FOR FIGURES

Figure 1. Map of the Ottawa River basin showing nuclear facility (CRNL and NPD) and sampling (Ottawa) locations.

Figure 2. Estimated contributions of fallout and CRNL radionuclides to the Ottawa River waters. The measured concentrations (Meyerhof, 1984; HWC, 1984-86) in the river waters are also shown.


Ontario
Quebec
QUEBEC
ONTARIO
Ottawa River
Rolphton
(NPD)
Chalk River
(CRNL)
Pembroke
Upper Basin
Lower Basin
Montreal
Ottawa
(Lemieux Island)
Quebec
New York
St. Lawrence River
UNITED STATES
0 20 40 60 80 100km
48°
47°
46°
45°
44°
81°
80°
79°
78°
77°
76°
75°
74°


Concentration (mBq L^{-1})
^{90}Sr
^{137}Cs
Measured
CRNL-Derived
Direct fallout over the river water surface
0
10
20
30
40
50
60
70
80
90
100
1960
1965
1970
1975
1980
1985



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