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TRANSPORT OF WEST VALLEY DERIVED RADIONUCLIDES TO LAKE ONTARIO VIA THE NIAGARA RIVER

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Implications/Executive Summary

The title study was conducted to study the possible migration of radioactive wastes stored at West Valley,NY toward Lake Ontario via the Niagara River.The data obtained shows that the controlled releases of the radionuclides are recorded in the deposited sediments. From these records it is inferred that (i) the maximum in Cs-137 activity in the core corresponds to 1969-70 and not the 1963 maximum due to atmospheric testing of nuclear weapons and (ii) once released the radioactive materials move toward Lake Ontario/Niagara River rather than disperse(and dilute further) in Lake Erie. The first result suggests that caution must be execised when dating the deposited sediments by the conventional Cs-137 technique.

The second result is of much more significance in that any accidental releases from the radioactive waste burial sites will move towards Niagara River/Lake Ontario and are likely to provide additional radiation dose to the public.The dose is likely to arise due to Sr-90 which would stay in the waters due to its conservative behaviour. My estimate (not given in the paper) is that during 1969-70(when the radioactivity levels in the receiving Cattaraugus Creek waters exceeded both the U.S.EPA and Nuclear Regulatory Commission(US) criteria)the controlled releases provided about .04 mrem of dose to the area residents in addition to about 0.2 mrem provided by Sr-90 due to weapons testing. The GLWQ Agreement objective for radioactivity is 1.0 mrem per year for all radioisotopes.Controlled amounts of radioactivity are still being released from the site which has been non-operational since 1972.These amounts are very small compared with earlier releases.Nevertheless, possibility always exists that radioactivity may be accidentally released from the burial trenches by overflow or by seepage of trench waters.(In 1976,rising waters in two trenches are known to have broken through their soil cover).The site has been monitored by several U.S. agencies in the past.Currently,only New York State Dept.of Environmental Conservation has a monitoring programme.The NYSDEC discontinued monitoring for the deadly plutonium isotopes in the recent past

MIGRATION DES RADIONUCLÉIDES DE WEST VALLEY VERS LE LAC ONTARIO PAR LE NIAGARA

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Incidences/Résumé administratif

On étudie la migration possible des déchets radioactifs stockés à West Valley (NY) vers le lac Ontario, par le Niagara. Les données obtenues montrent que les rejets contrôlés de radionucléides se retrouvent dans les dépôts de sédiments. On déduit de l'examen des sédiments (i) que l'activité maximale de Cs-137 selon les carottes correspond à 1969-1970 et non pas au maximum de 1963, conséquence des essais atmosphériques d'armes atomiques et (ii), qu'une fois libérées, les substances radioactives se dirigent vers le lac Ontario par le Niagara et ne se dispersent pas (pour ensuite se diluer) dans le lac Érié. Le premier résultat indique que nous devons faire preuve de prudence lorsque l'on applique la technique classique du Cs-137 pour la datation des dépôts de sédiments.

Le deuxième est beaucoup plus important car il montre qu'en cas de rejet accidentel dans les sites d'enfouissement de déchets radioactifs, les substances libérées s'écouleraient dans le Niagara puis dans le lac Ontario et viendraient probablement augmenter la dose des radiations auxquelles le public est exposé. Cette augmentation est probable car le Sr-90 resterait dans l'eau. Selon mes propres estimations, qui ne figurent pas dans le document, en 1969-1970 lorsque les niveaux de radioactivité dans les eaux réceptrices de la Cattaraugus ont dépassé les critères de l'EPA et de la Nuclear Regulatory Commission des É.-U., les rejets contrôlés constituaient environ 0,04 mrem de la dose à laquelle étaient exposés les habitants de la région, outre environ 0,2 mrem pour le Sr-90 provenant des essais d'armes. Selon l'accord sur la QEGL, l'objectif de radioactivité est de 1 mrem par an pour tous les isotopes radioactifs. Les rejets contrôlés d'éléments radioactifs se poursuivent au site en question qui n'est plus en service depuis 1972. Les quantités libérées sont minimes par rapport aux rejets antérieurs. La possibilité d'un rejet accidentel demeure cependant, soit par débordement soit par suintement des eaux des tranchées d'enfouissement. (On sait qu'en 1976 les eaux ont débordé à la surface de deux tranchées, franchissant la couche de sol de couverture.) Plusieurs organismes des É.-U. ont surveillé le site mais à l'heure actuelle seul le New York State Department of Environmental Conservation se charge de cette surveillance. Récemment cet organisme a arrêté la surveillance des isotopes meurtriers de plutonium.

The pollution of the Niagara River/Lake Ontario region is a topic of major importance. Several studies have defined the salient features of area pollution by toxic organic compounds[1-3] and heavy metals[4,5]. The existence of several nuclear facilities in the drainage basin[6] leads to the possibility of contamination of this area by radioactive materials as well. By far the most toxic radioactive materials are stored at the Western New York Nuclear Service Center (WNYNSC), located at West Valley, N.Y., U.S.A. The site comprises of a shutdown facility for the reprocessing of spent nuclear fuel, a storage facility for high-level radioactive liquid wastes, a treatment facility for low-level liquid wastes, and burial areas for low- and intermediate-level radioactive solid wastes. Between 1969 and 1972, nearly 13 TBq of liquid radioactive waste, excluding 550 TBq of liquid tritium, were discharged to the local drainage system[7]. The resulting average ^{90}Sr levels in the adjoining Cattaraugus Creek, which empties into Lake Erie, for 1969-71 exceeded both the U.S. Environmental Protection Agency standard for drinking water and the U.S. Nuclear Regulatory Commission's technical specifications for the creek[8]. In 1978, besides 164 tonnes of uranium in the spent fuel storage facility, nearly 2.3 EBq of short-lived and 0.9 PBq of long-lived radioactive wastes were variously located at the site[6]. Although no fuel has been reprocessed since 1975, releases of controlled amounts of radioactive wastes to the local drainage system have continued[9]. Besides controlled releases, the possibility also exists that

radioactivity may be released to the local aquatic ecosystem directly from the burial trenches by overflow or by seepage of trench waters. Indeed, in 1976, rising waters in two trenches had broken through their soil cover[10]. The results presented in this communication show that the West Valley-derived radionuclides are quickly transported to Lake Ontario via the Niagara River.

Water and sediment samples, collected from locations shown in Fig. 1, were analysed by high-resolution gamma-ray spectrometry as described earlier[11,12]. Surficial (~1 cm) sediments from all five locations in Lake Ontario were also analysed for ^{238}Pu and $^{239,240}Pu$ as described elsewhere[13]. A sediment core from station 207 was also raised in September 1982 and 1-cm segments assayed for ^{137}Cs, ^{210}Pb and ^{226}Ra by gamma-ray spectrometry. The ^{210}Pb dates were assigned using the procedures described by Durham and Oliver[2].

The data presented in Table 1 shows that several radionuclides, generally not associated with the routine nuclear fallout in the study area[11], are present in the Lake Ontario surface sediments. The activity ratios of common fallout radionuclides ^{137}Cs, ^{238}Pu, $^{239,240}Pu$ and ^{241}Am (Table 2) which are also released in nuclear fuel reprocessing operations further sugggest non-fallout inputs. The fallout activity ratios of $^{137}Cs/^{239,240}Pu$ and post-SNAP 9A $^{238}Pu/^{239,240}Pu$ in this latitudinal band (40-50°N) are reckoned to be about 67 and 0.04, respectively[14,15]. The $^{241}Am/^{239,240}Pu$ activity ratio in recent fallout samples would be very close to zero

since ^{241}Am (half-life 433 y) is generated only from ^{241}Pu (half-life 14.4 y). Assuming a fallout $^{241}Pu/^{239,240}Pu$ activity ratio of 14.0 in fresh debris[16], a stratospheric residence time of one year for the debris, and a sedimentation rate of 1 cm/y at a location such as station 207, the fallout $^{241}Am/^{239,240}Pu$ activity ratio would be only about 0.04. The data given in Table 2, especially the activity ratios for the transuranics where stratospheric and/or sedimentary fractionation and diffusion in the sediments are least likely to affect the ratios, clearly suggest non-fallout inputs of these radionuclides in the study area.

Since the Niagara River supplies almost all the sediment at sampling locations in Lake Ontario[17], two water sampling stations were established in the river to trace the source of these radionuclides. Table 3 gives the results of gamma-ray spectrometric measurements on these water samples. The results clearly show that the same non-fallout radionuclides are invariably detected in the water samples as are present in the Lake Ontario sediments. All these radionuclides have been previously[18] detected at various sampling sites in the Cattaraugus Creek. The detection of relatively short-lived radionuclides ^{54}Mn, ^{65}Zn and ^{106}Ru in the present investigation is unexpected, if not impossible, considering that the WNYNSC has not received any new wastes since 1975. It may be suggested that these short-lived radionuclides originate from the State University of New York at Buffalo research reactor wastes, controlled amounts of which

are discharged into the municipal sanitary sewer system[19] that eventually releases post-treatment effluents into the Niagara River. Two arguments are used to discount this possibility. Firstly, the available data[20] suggests that these radionuclides will be quantitatively retained on the sewage treatment plant sludge and not released with the plant effluent. The non-detection of any medically-useful radionuclides in the present study lends further support to this argument. Secondly, unlike ^{54}Mn and ^{65}Zn which may be routinely produced by (n,Y) reactions on nuclei present in the reactor pool water, the fission product ^{106}Ru will be present in the pool water only if fissile material is exposed to water. Such a situation occurred in January 1971 when the cladding on a fuel element failed[21]. Subsequent gamma-spectrometric measurements showed the presence of several radionuclides in the reactor pool water except for ^{106}Ru (fission yield, 0.39%) although ^{103}Ru (fission yield, 3.1%) was easily detected[21,22]. It is also worth noting that both studies failed to detect any ^{137}Cs in the reactor pool water.

The supporting evidence as to the source of non-fallout radionuclide inputs to Lake Ontario is provided by an analysis of the ^{137}Cs profile (Fig. 2) of a sediment core retrieved from station 207. Assignment of the ^{210}Pb-derived dates to the core sections reveals that the maximum ^{137}Cs deposition at this location occurred around 1969-70. This corresponds well with the available data[8,9] which shows that the levels of West Valley effluents in Cattaraugus Creek,

monitored before and after the 1966 start-up of fuel reprocessing plant, started increasing during 1967 and peaked in 1970.

Accepting that the sole source of non-fallout inputs is the West Valley effluent and that the dominant flow of Lake Erie into the Niagara River carries all suspended inputs from Cattaraugus Creek along the direction of the flow, the deposition of ^{137}Cs in Lake Ontario during 1967-82 may be estimated. In view of the unavailability of precise release data for this period, the estimates of ^{137}Cs concentration are based on the measured ^{90}Sr levels (fallout plus West Valley-delivered) in Cattaraugus Creek and a $^{137}Cs/^{90}Sr$ activity ratio of 1.5. This activity ratio is indicated by the limited release data[7] for 1971 and 1972 and is similar to that used for fallout studies.

From the flow characteristics of the Cattaraugus Creek[10] (20.7 m^3/s) and the Niagara River[5] (5500 m^3/s) and an inferred total (i.e. dissolved plus particulate) ^{137}Cs concentration of 1.22 Bq/L for 1967 in the Cattaraugus Creek waters, a ^{137}Cs concentration of about 4.5 mBq/L is derived at the confluence of the Niagara River and Lake Ontario. The particulate fraction concentration of ^{137}Cs is estimated to be about 460 mBq/g using a distribution coefficient (K_d) of 10^5. This value of K_d is estimated from the data given by Onishi et al.[18] for dissolved and particulate ^{137}Cs in the Cattaraugus Creek waters during 1978-79, and is similar to the one estimated by Alberts and Muller[23] for Lake Michigan sediments. The ^{137}Cs input per unit area

at this station, 138.3 mBq cm^{-2}, is then obtained by multiplying particulate ^{137}Cs concentration by the estimated mass sedimentation rate (0.279 g cm^{-2} y^{-1}) for 1967. Numerical integration of decay-corrected (to September 1982) estimates of annual deliveries of West Valley-derived ^{137}Cs, shown in Fig. 2 using an arbitrary scale along with estimates of atmospheric fallout[24], shows that the West Valley-delivered ^{137}Cs (968 mBq cm^{-2}) accounts for about 67% of the total ^{137}Cs inventory (1.449 Bq cm^{-2}) at this location. Fallout ^{137}Cs (395 mBq cm^{-2}) accounts for a further 27% of the total inventory. The balance, 86 mBq cm^{-2} or about 22% of the fallout inventory, must then derive from Lake Erie. Indeed, Edgington and Karttunen[25] have estimated that about 25% of the direct fallout ^{137}Cs in Lake Erie is not deposited in bottom sediments.

The foremost implication of the present study is that any accidental releases of radionuclides from the WNYNSC are likely to move toward the Niagara River and then Lake Ontario rather than disperse in Lake Erie. While many radionuclides may be removed on water filtration plant floc[26], conservatives such as ^{90}Sr will probably pass into the drinking water, thus providing most of the radiation dose to the area population.

An important corollary to this investigation provides that the peak ^{137}Cs activity in the area sediments corresponds to the 1970 discharges from the WNYNSC and not the 1963 fallout maximum as has been assumed in a recent sedimentological study[27]. Presumably, this

feature is retained in the major depositional basins of Lake Ontario as well since the Niagara River, supplying nearly 50% of the total sediment input to the lake, is thought to affect the whole lake[17]. The influence of West Valley effluents on the Niagara Basin (station 206) is evident from the data given in Tables 1 and 2.

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Table 1 Levels (mBq/g dry) of radionuclides in the surface sediments of Lake Ontario at the mouth of Niagara River (1982)

Radionuclide	Half-Life	Station				
		206	207	208	209	210
^{54}Mn	312 d	ND	0.4±0.3	1.5±0.4	ND	0.5±0.4
^{60}Co	5.27 y	2.9±0.6	1.6±0.5	0.8±0.6	0.5±0.4	ND
^{65}Zn	244 d	ND	1.2±0.9	ND	ND	0.8±0.7
^{106}Ru	368 d	6.2±4.0	ND	8.0±3.8	4.6±3.4	ND
^{125}Sb	2.77 y	8.5±1.4	3.2±0.9	2.2±0.9	2.3±0.9	ND
^{134}Cs	2.06 y	8.3±4.1	ND	ND	5.3±3.2	7.0±2.9
^{137}Cs	30.1 y	256.0±2.1	109.2±1.1	77.3±1.0	76.3±1.1	79.3±1.1
^{144}Ce	284.2 d	11.2±2.0	12.2±1.3	11.3±1.6	7.2±1.4	4.5±1.4
^{155}Eu	4.96 y	10.4±1.0	6.4±0.7	5.2±0.7	2.9±0.7	3.4±0.7
^{207}Bi	38 y	ND	ND	ND	0.7±0.5	ND
^{238}Pu	87.8 y	0.06±0.01	0.23±0.02	0.05±0.01	0.27±0.01	0.07±0.01
$^{239,240}Pu$	2.4×10^{4} y, 6537 y	3.33±0.08	1.15±0.03	0.75±0.02	0.74±0.02	0.90±0.01
^{241}Am	433 y	1.1±0.6	0.5±0.3	ND	ND	0.6±0.3

Errors are based on 1σ counting statistics; ND, not detected.

Table 2 Activity ratios of some radionuclides in surface sediments of Lake Ontario at the mouth of Niagara River

Radionuclide	Station				
	206	207	208	209	210
$^{137}Cs/^{239,240}Pu$	76.9	95.0	103.1	103.1	88.1
$^{238}Pu/^{239,240}Pu$	0.02	0.20	0.07	0.36	0.08
$^{241}Am/^{239,240}Pu$	0.3	0.4	No data	No data	0.7

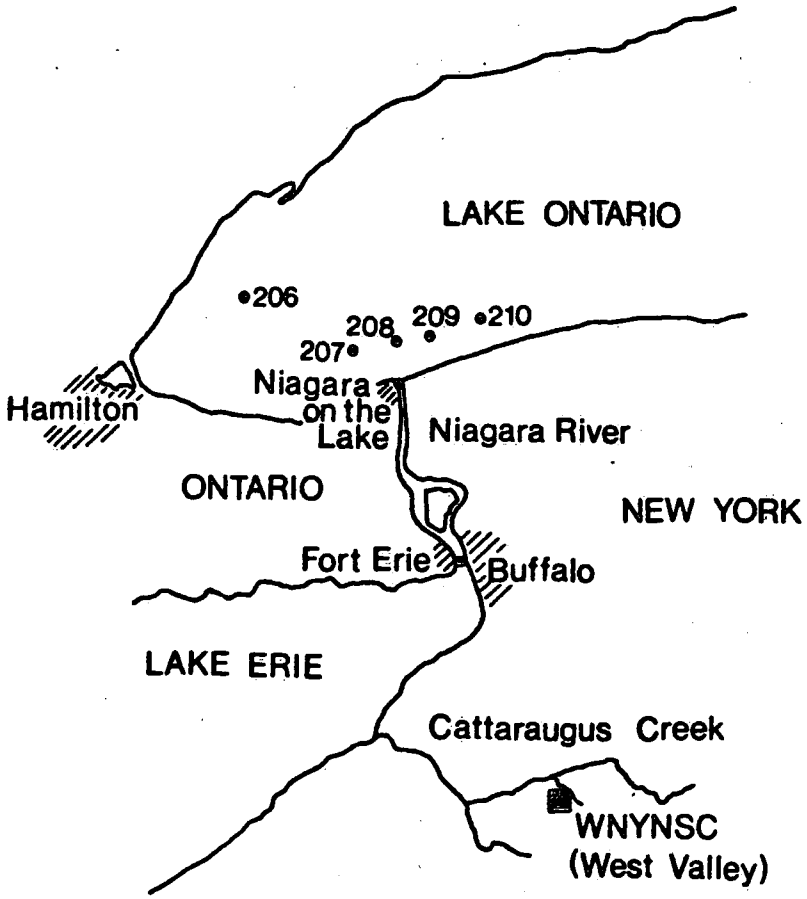
Table 3 Levels (mBq/L) of non-fallout gamma-emitters in the Niagara River surface waters

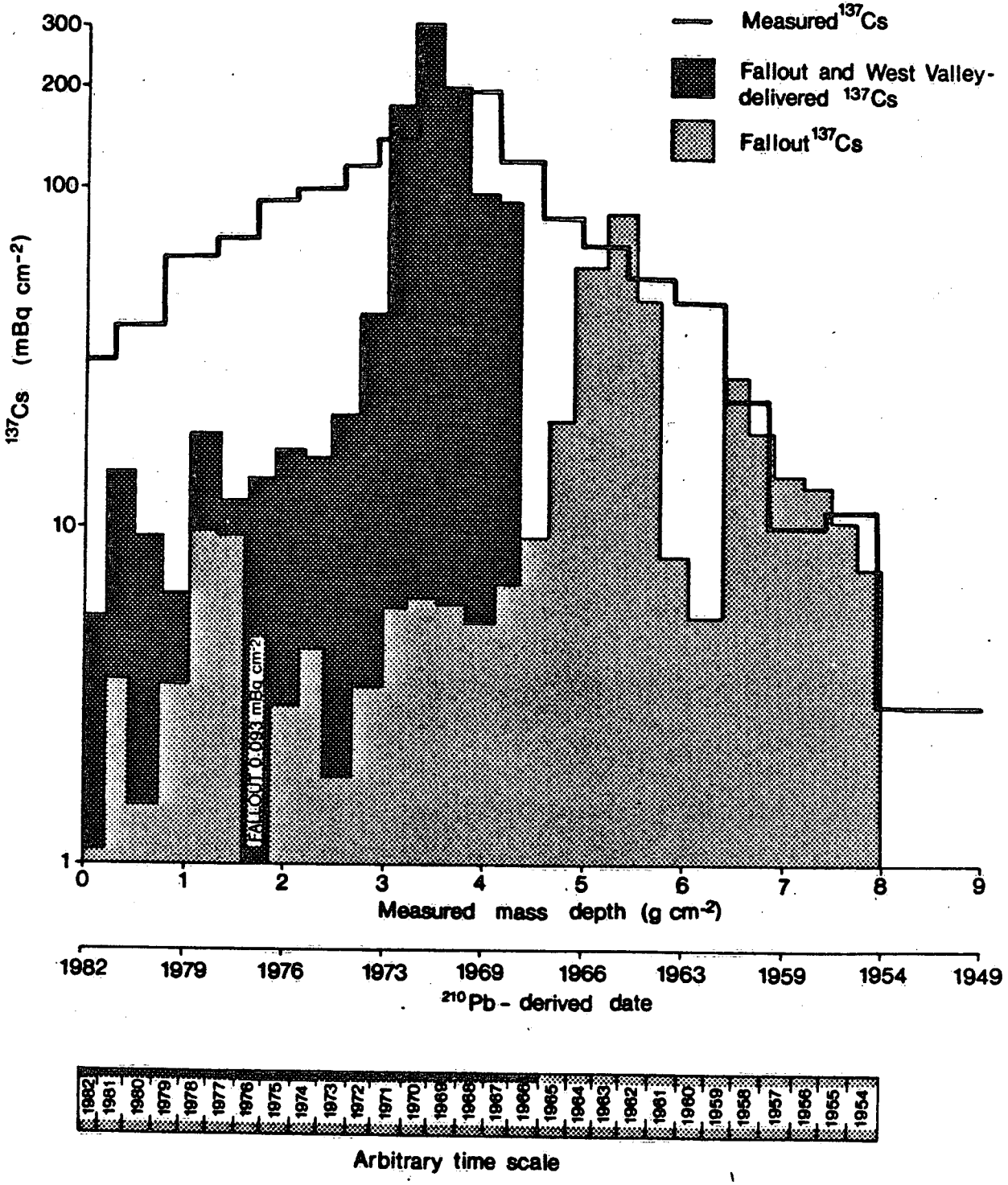
Location	Year, Quarter	^{54}Mn	^{60}Co	^{65}Zn	^{106}Ru	^{134}Cs
Niagara-on-the-Lake	1982, 1-2Q	ND	ND	11.2±1.6	ND	9.5±5.2
	3Q	ND	ND	3.2±0.4	ND	ND
	4Q	ND	0.2±0.1	0.9±0.3	ND	ND
Niagara-on-the-Lake	1983, 1Q	0.3±0.2	ND	2.0±0.6	ND	ND
	2Q	0.4±0.2	0.5±0.3	0.8±0.5	ND	ND
	3Q	ND	0.3±0.2	1.3±0.6	ND	ND
	4Q	ND	ND	ND	ND	ND
Fort Erie	4Q	ND	0.5±0.3	1.0±0.5	ND	ND
Niagara-on-the-Lake	1984, 1Q	ND	ND	ND	ND	ND
	3Q	ND	0.8±0.3	1.3±0.7	2.6±1.4	ND
	4Q	ND	0.3±0.2	ND	ND	ND
Fort Erie	1Q	ND	0.3±0.2	ND	2.4±1.7	ND
	2Q	ND	ND	ND	3.1±2.0	ND
	3Q	ND	ND	ND	4.1±2.7	ND
	4Q	ND	ND	ND	ND	

Errors are based on 1σ counting statistics; ND, not detected.

Figure Captions

1. Map showing locations of sampling sites.
2. ^{137}Cs profile in ^{210}Pb-dated sediment core from station 207. The shaded areas, corresponding to an arbitrary time scale, depict inputs of fallout and West Valley-delivered ^{137}Cs.


LAKE ONTARIO
206
208
209
210
207
Niagara on the Lake
Hamilton
Niagara River
ONTARIO
NEW YORK
Fort Erie
Buffalo
LAKE ERIE
Cattaraugus Creek
WNYNSC
(West Valley)


Measured ^{137}Cs
Fallout and West Valley-delivered ^{137}Cs
Fallout ^{137}Cs
^{137}Cs (mBq cm^{-2})
300
200
100
10
1
FALLOUT 0.093 mBq cm^{-2}
0 1 2 3 4 5 6 7 8 9
Measured mass depth (g cm^{-2})
1982 1979 1976 1973 1969 1966 1963 1959 1954 1949
^{210}Pb - derived date
1982 1981 1980 1979 1978 1977 1976 1975 1974 1973 1972 1971 1970 1969 1968 1967 1966 1965 1964 1963 1962 1961 1960 1959 1958 1957 1956 1955 1954
Arbitrary time scale