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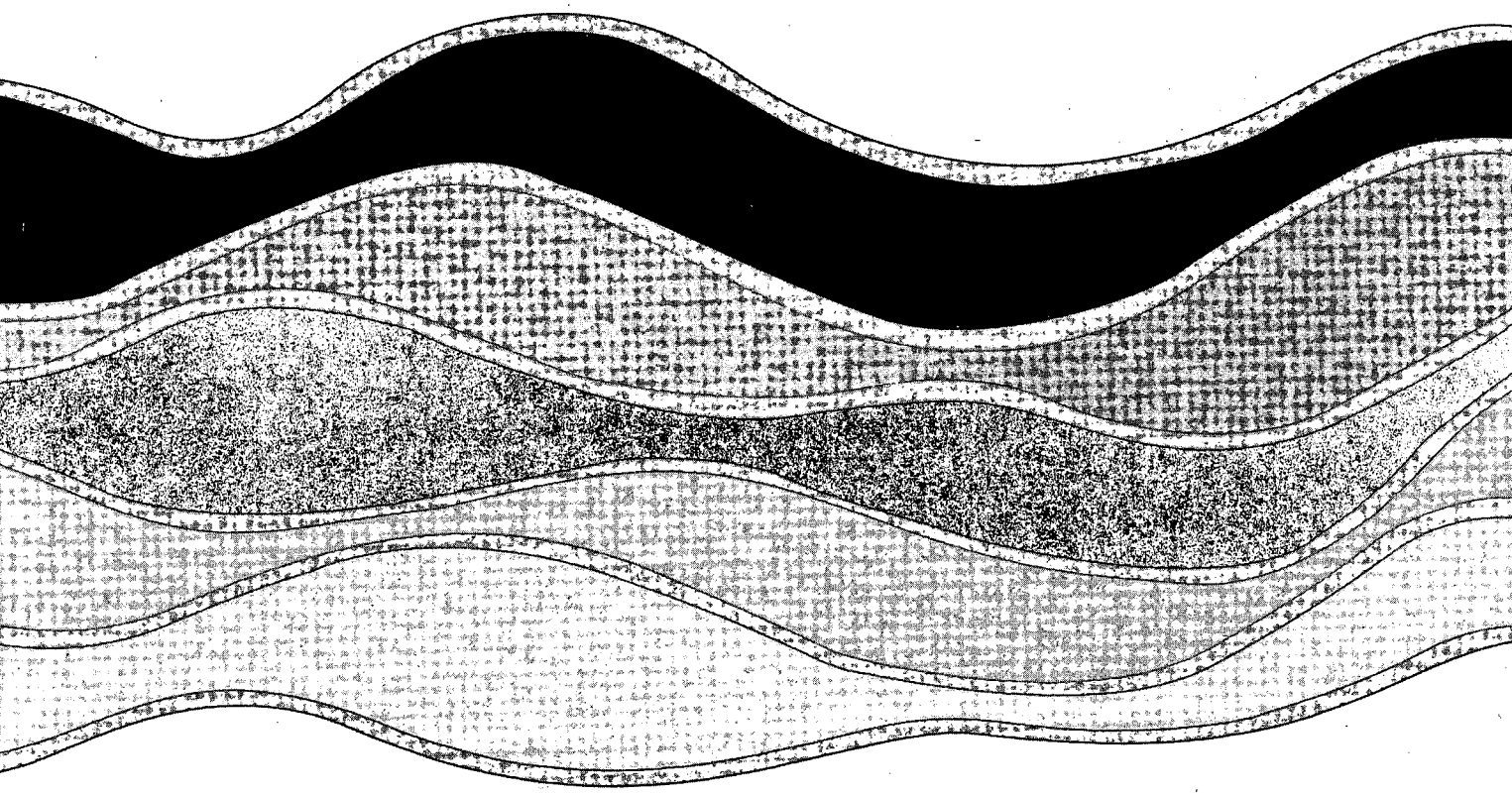

RADIOACTIVITY IN THE GREAT LAKES

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NWRI Contribution No. 91-03

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MANAGEMENT PERSPECTIVE

This review constitues the first comprehensive survey of radioactivity in the Great Lakes. Aspects considered include sources, ecosystem distribution and radiation dose to humans and aquatic biota. All reported findings are critically assessed and the adequacy of applicable guidelines and criteria examined. Refined estimates of fallout radionuclide deposition for each lake are also reported.

Nuclear reactor sites, a former nuclear fuel reprocessing area, and the uranium mine waste areas are by far the most significant potential sources of release of radioactivity to the aquatic system. The present levels of man-made radionuclides in ambient waters, biota and sediments are, however, almost exclusively derived from previous fallout from nuclear weapons testing, and provide very little radiation dose to area residents consuming lake water. The existing models adequately describe the interlake transport of radioactivity though some refinement is needed. Studies on the dispersal of radioactive pollutants are very limited. The role of chemical parameters in the radionuclide cycling has been extensively investigated in Lake Michigan, while studies on biological uptake and dosimetry are very limited. The review outlines several areas requiring further research and increased monitoring efforts to develop a

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better understanding of the impact of radioactive releases. These include: studies on the sources and dispersal of tritium, studies on the behaviour of ^{99}Tc (a highly toxic nuclear fission product for which no data are available), and increased water and fish monitoring for transport model verification and dosimetric purposes.

The review also demonstrates a need for developing a radiological objective for fish the consumption of which may impart more radiation dose (at least to some critical groups) than the consumption of lakewater. It is also suggested that provincial guidelines for the open water disposal of dredged spoils be upgraded to include radioactive substances.

PERSPECTIVES DE LA DIRECTION

Ce tour d'horizon est la première étude complète de la radioactivité dans les Grands Lacs. Il y est question entre autres des sources, de la distribution dans l'écosystème et de la dose d'irradiation pour l'humain et les organismes du milieu aquatique. On y fait l'analyse critique de toutes les observations présentées ainsi que l'évaluation de la justesse des directives et critères applicables. Enfin, on donne une estimation précise du dépôt de radionucléides dans chacun des lacs.

Les réacteurs nucléaires, une ancienne installation de retraitement des combustibles nucléaires et des décharges de déchets de mines d'uranium sont de loin les plus importantes des sources possibles de radioactivité dans le milieu aquatique. Les concentrations actuelles de radionucléides d'origine anthropogène dans les eaux, le biote et les sédiments sont toutefois presque exclusivement imputables aux retombées qu'ont entraînées les essais d'armes nucléaires faits antérieurement et ne représentent qu'une très petite dose d'irradiation pour les habitants qui consomment l'eau des lacs. Les modèles actuels donnent une description adéquate du transport des matériaux radioactifs d'un lac à l'autre, mais il y aurait place pour certains raffinements. Les études portant sur la dispersion des polluants radioactifs sont très limitées. On a étudié de façon poussée le rôle des paramètres chimiques dans le cycle des radionucléides dans le lac Michigan; par contre il s'est fait très peu de choses sur l'absorption biologique et la dosimétrie. L'analyse fait ressortir divers points sur lesquels il y aurait lieu de pousser la recherche et les activités de surveillance pour mieux comprendre les conséquences des rejets radioactifs. Il s'agit notamment des aspects suivants : étude des sources de tritium et de la dispersion de cet isotope, étude du comportement du ^{99}Tc (un produit de fission nucléaire très toxique sur lequel on ne possède aucune donnée) et intensification

de la surveillance de l'eau et des poissons pour la vérification des modèles de transport et pour des usages en dosimétrie.

L'analyse révèle aussi qu'il y aurait lieu d'établir un objectif de radioactivité applicable aux poissons dont la consommation représente une dose d'irradiation plus élevée (du moins dans le cas de certains groupes critiques) que la consommation de l'eau des lacs. On propose également d'étendre les directives provinciales sur l'élimination des déblais de dragage en eau libre pour y inclure les substances radioactives.

ABSTRACT

Studies of radioactivity in the Laurentian Great Lakes are reviewed to evaluate the impact of radionuclide dissemination on the world's foremost freshwater aquatic ecosystem. The status of radiologically degraded areas is also reported. Significant amounts of radioactivity are stored in the basin which has numerous nuclear reactors as well as uranium mine waste areas. The prevailing low levels of artificially-produced radionuclides, arising largely from previous fallout inputs, provide very little radiation dose to the area residents consuming lake water. The interlake transport of radionuclides is adequately described by existing models though some refinement of the source term is needed. Revised estimates of fallout over each lake are given, but no data are available to estimate drainage basin contributions. Only limited information is available on the dispersal of radioactive pollutants. The influence of chemical parameters on the radionuclide cycling has been extensively investigated in Lake Michigan and, to a lesser degree, in Lake Ontario. The need for developing a radiological objective for fish becomes apparent from an assessment of the very few data collected thus far on the biological and dosimetric aspects. Several research and monitoring needs are also identified.

RÉSUMÉ

On fait l'analyse des études portant sur la radioactivité dans les Grands Lacs laurentiens pour évaluer les conséquences de la dispersion des radionucléides dans l'écosystème d'eau douce le plus important du monde. On décrit également l'état des zones touchées par la radioactivité. Des quantités significatives de substances radioactives sont conservées dans le bassin des Grands Lacs, région où il existe de nombreux réacteurs nucléaires ainsi que des installations d'élimination des déchets de mines d'uranium. Les radionucléides produits artificiellement, provenant en grande partie des retombées qui ont eu lieu avant l'aménagement de ces installations, sont présents en concentrations généralement faibles et ne représentent qu'un très petite dose d'irradiation pour les habitants de la région qui consomment l'eau des lacs. Le transport des radionucléides d'un lac à l'autre est adéquatement représenté par les modèles actuels, mais il y aurait lieu de raffiner davantage le terme représentant les sources. On donne des estimations revues des retombées dans chaque lac; aucune donnée ne permet toutefois d'estimer l'apport du bassin hydrographique. On ne possède que des données limitées sur la dispersion des polluants radioactifs. L'influence des paramètres chimiques sur le cycle des radionucléides dans le lac Michigan a fait l'objet d'une étude poussée; on a examiné le même phénomène dans le lac Ontario, mais de façon moins approfondie. L'évaluation des rares données recueillies jusqu'ici sur les aspects biologiques et dosimétriques révèle qu'il y aurait lieu d'établir un objectif de radioactivité applicable aux poissons. On mentionne également divers points au sujet desquels il serait bon de pousser la recherche et les activités de surveillance.

INTRODUCTION

Man's mastery of the atom began in 1942 when, for the first time, a self-sustaining nuclear chain reaction was successfully triggered and controlled at the Chicago Pile in the Great Lakes basin. Subsequent military and civilian applications of nuclear energy have released significant amounts of artificially-produced radioactivity to the global environment. These releases have undoubtedly added to the levels of the natural radiation, and concern has been expressed for the welfare of life exposed to this additional radiation.

An area of particular interest is the five Great Lakes, the largest and most valuable assemblage of freshwater resources in the world. The long water residence times in the upper Great Lakes ensure that some of the long-lived fallout radionuclides will continue to persist in this intricate ecosystem for many years to come. The presence of significant uranium deposits in the Great Lakes basin and abundant water supply has attracted nuclear power industry to the shores of all the lakes (except Lake Superior) to support the energy needs of other industries and area population. Both planned and unplanned releases of radioactivity from nuclear fuel cycle operations impact the Great Lakes ecosystem. In addition to these two major sources, the

Great Lakes also receive relatively minor inputs of radioactivity from other sources.

Recent investigations of the impact of radionuclides in the Great Lakes have included: monitoring of nearshore and open waters, sediments and biota; transport behaviour and cycling studies; and studies on the forms and modes of association. The present review thus examines the currently available information on radionuclides of concern with the view of providing a perspective on their relative inputs, pathways and persistence in the Great Lakes. An attempt is also made to identify gaps in existing knowledge on the fate and effects of such radionuclides in the Great Lakes.

SOURCES OF RADIOACTIVITY TO THE GREAT LAKES

Radionuclides can enter the Great Lakes ecosystem as a result of both natural and man-made processes. The principal natural processes which introduce radioactivity are the weathering of rocks which contain uranium- and thorium-series radionuclides, and fallout of such cosmic ray-produced radionuclides as ^{3}H, ^{7}Be, and ^{14}C.

The Great Lakes basin is unique in that it contains nearly all components of the nuclear fuel cycle activities (Fig. 1). This has resulted in technologically-enhanced natural radiation levels through such activities as uranium mining, milling, conversion and fuel fabrication, and release of artificially-produced radionuclides through nuclear power reactors and the nuclear fuel reprocessing plant (closed since 1972) at West Valley, NY. Additional inputs of both types of radioactivity have been provided by the waste management facilities. Minor inputs of radioactivity result from medical uses of radioisotopes (Durham and Joshi, 1979) and from coal-fired electrical generating plants (IJC, 1983). Limited available data (NYSDH, 1986-88) suggests that the research and industrial applications of radioisotopes are likely to constitute only a very minor source of anthropogenic radioactivity to the Great Lakes. The recently-commissioned (1989) tritium removal facility at Darlington could be another significant source of this radionuclide in Lake Ontario.

Fallout from nuclear weapons testing has been a major source of anthropogenic radioactivity in the Great Lakes ecosystem. The first injection of nuclear fission and activation products to the atmosphere took place in 1945. Regular atmospheric testing of nuclear weapons started in 1952, increased significantly in the late 1950s, and peaked in 1963 when a partial test ban treaty

came into force. Since then only occasional testing has been carried out in the northern hemisphere. Fallout from the Chernobyl nuclear accident deposited negligible amount of long-lived radioactivity in the area (Joshi, 1988a).

Radionuclides from nuclear facilities may enter the Great Lakes basin via both atmospheric and liquid emissions. For naturally-occurring radionuclides, ^{222}Rn (and its progenies) from uranium mining and milling locations, and atmospheric particulate emissions from uranium refining industry and coal-fired plants are the more significant components of atmospheric delivery. However, on a basinwide basis, their contribution is very small. Besides nuclear weapons testing fallout, anthropogenic radionuclides are also released to the atmosphere during nuclear power production and nuclear fuel reprocessing activities. Most of this radioactivity is comprised of short-lived inert gases, but ^{3}H (half-life 12.3y) and particle-reactive ^{129}I (half-life 1.7 x 10^{7}y) and ^{131}I (half-life 8.1d) may be released in measureable concentrations. Of these, ^{129}I was largely released from the West Valley spent-fuel reprocessing plant during 1966-72. Atmospheric emissions of other long-lived and particle-reactive anthropogenic radionuclides comprise a very small fraction of the total release (UNSCEAR, 1977). In all, with the exception of tritium, anthropogenic radionuclides from nuclear facilities are released to the system via liquid effluents.

RELATIVE SIGNIFICANCE OF SOURCES OF RADIONUCLIDES

Naturally-Occurring Radionuclides

All uranium mining and milling activities in the Great Lakes basin are located in Canada. Commencing in the mid 1950s, these operations have generated about 100 mt of wastes in the Elliot Lake and Bancroft areas; the Agnew Lake mine uses a solution mining process and, therefore, does not produce conventional tailings. Based on the available analytical data (IJC, 1979), the former two areas are estimated to contain about 2 PBq of ^{226}Ra (half-life 1620y), the most toxic of the radionuclides present in the wastes. Because of the presence of precursor ^{230}Th (half-life 8 x 10^4y), the ^{226}Ra activity in the tailings will remain for thousands of years.

The subsequent refining of the yellowcake at Port Hope has generated about 25 TBq of ^{226}Ra most of which was deposited at the Port Granby radioactive waste management site from 1955 to 1977 (Platford et al., 1984). Much smaller quantities of these wastes were released to the Port Hope Harbour or deposited at the Welcome waste management site. The 1983 relocation of the Port Hope UO_3 plant to Blind River has removed the source of ^{226}Ra to the harbour which, however, still retains sediments contaminated

with ^{226}Ra (and other radionuclides) from refining and earlier radium (1933-1952) and uranium (1952-1955) recovery operations. Fuel fabrication facilities release even smaller amounts of ^{226}Ra to the environment (IJC, 1979). Taken together, more than 98% of the ^{226}Ra activity originally present in the ores is retained in the mining and milling wastes which are, therefore, the dominant source of technologically-enhanced natural radiation in the Great Lakes basin. The waste management sites at Lewiston and West Valley in the U.S. also contain small amounts of naturally-occurring radionuclides including ^{226}Ra.

Artificially-Produced Radionuclides

The first nuclear power reactor in the Great Lakes basin became operational in 1963, the year of maximum fallout activity. Since then while the fallout in the area from weapons testing has sharply declined, the nuclear generating capacity along the shores of the Great Lakes has increased by over two orders of magnitude (Fig. 2). Both these sources generate same radionuclides albeit in somewhat different proportions. Typical amongst these are ^{90}Sr (half-life 29y) and ^{137}Cs (half-life 30.1y).

The deposition of fallout ^{90}Sr has been measured at several locations in and around the Great Lakes basin (NRCC, 1983; Larsen, 1985). Such information is of use in deriving estimates

of radiation dose to the public and in modelling the long-term behaviour of radionuclides in waters and bottom sediments. The deposition of fallout radioactivity is expected to show latitudinal dependence (Hardy et al., 1973; UNSCEAR, 1982; Larsen, 1985) with deposition declining in northerly latitudes. This trend is clearly discernible in Fig. 3 where cumulative (1959-76 inclusive) deposition of ^{90}Sr per unit area (NRCC, 1983; Aarkrog et al., 1989) is plotted as a function of latitude. It is evident from Fig. 3 that no single value may be used for estimating fallout over the five Great Lakes. A regression analysis of the measured deposition of fallout ^{90}Sr at various Great Lakes locations over the time period spanning maximum fallout shows that the flux F of this radionuclide at a Great Lakes latitude N with reference to that at New York City (for which location most consistent data are available to us) may be given by the equation:

$$F_N = F_{NYC} \times (-7.9 \times N + 662.23)/456.6 \qquad (1)$$

The values of fallout ^{90}Sr flux at a mid-basin location for each of the Great Lakes are given in Appendix 1. Estimates of ^{137}Cs are also given in Appendix 1. A $^{137}Cs/^{90}Sr$ activity ratio of 1.6 (NRCC, 1983) was used in deriving these estimates though there is some evidence (Sherrill et al., 1975) that the ratio may vary considerably.

Estimates of cumulative inputs of fallout ^{90}Sr and ^{137}Cs, derived by using data in Appendix 1 and the water surface areas (CCGLBHHD, 1977) of different lakes are shown in Figs. 4 and 5, respectively. Undoubtedly, some fraction of the radioactivity falling over land is also removed to the lakes; however, no data are available to estimate these inputs. Available data on other watersheds (Ritchie and McHenry, 1990) indicate that very little ^{137}Cs is removed by erosional processes. Our own measurements (S.R. Joshi, B.S. Shukla and R.C. McCrea, unpublished data) on the adjoining Ottawa River watershed show that this radionuclide has a mean residence time of about 8000y in the area soils. The conservative chemical behaviour of ^{90}Sr, on the other hand, allows this radionuclide either to migrate deeper into the soils (Walton, 1963) or to be removed to the waterbody. Therefore, the contribution of fallout ^{90}Sr to the lakes is likely underestimated, perhaps by up to 0.8% of land surface input per year if the radionuclide's transport pattern is analogous to that in smaller watersheds (Menzel, 1974). Estimates for fallout ^{3}H inputs to each of the lakes were also derived from published information (UNSCEAR, 1977) and are shown in Fig. 6.

The releases of the three radionuclides from nuclear facilities in the basin are also shown in Figs. 4-6. Only discharges via liquid effluents were considered as airborne particulate emissions contain exceedingly small amounts of ^{90}Sr

and ^{137}Cs (UNSCEAR, 1977). Atmospheric releases of ^{3}H from heavy water reactors are significantly higher than those via the liquid effluents (UNSCEAR, 1977, 1982 and 1988). No data are available to estimate either the fraction of atmospheric release that will be transported to the lakes or the fraction of tritium lost to the atmosphere from the lakes via evaporation. Consequently, the values shown in Fig. 6 should be regarded as approximate though the present estimates are in general agreement with the earlier projections by Gustafson (1970) and Sullivan and Ellett (1977). Since most of the spent fuel is retained at the nuclear generating stations (IJC, 1979), estimates of stored ^{90}Sr and ^{137}Cs are also shown in Figs. 4 and 5, respectively. Radionuclide release and production estimates were inferred from a variety of sources (AECB, 1987; Eichholz, 1983; Gillespie et al., 1984; IJC, 1977, 1978, 1979, 1983 and 1989; Mehta, 1982; UNSCEAR, 1977, 1982 and 1988). The values shown are cumulative from the start-up date to 1989. Estimates for Lake Erie also include contributions from the nuclear fuel reprocessing activities.

An inspection of the data given in Figs. 4-6 shows that fallout is by far the more dominant source of these radionuclides in the Great Lakes waters. The near-exceptions include ^{137}Cs in Lakes Ontario and Michigan and ^{3}H in Lake Huron. This reflects relatively large releases of ^{137}Cs from boiling water reactors and of ^{3}H from heavy water reactors. The relatively higher inputs of

facility-derived ^{90}Sr in Lake Erie are due to contributions from the nuclear fuel reprocessing plant. Taken together, the data show that the prevailing levels of these radionuclides in the Great Lakes are essentially due to fallout inputs. However, the concern for the radioactive contamination of this ecosystem derives from any accidental discharge of radioactivity from an operating nuclear reactor or from a storage facility in the basin. The stored radioactivity already exceeds combined fallout and planned releases by several orders of magnitude (Figs. 4 and 5) and will continue to increase in the foreseeable future. Even though the prevailing levels of radionuclides in the ecosystem may not have serious human health implications, continuing investigations of their interaction and transport in various compartments are required to develop a sound basis for handling any possible accidental release of radioactivity.

CHEMICAL FORMS AND INTERACTIONS OF RADIONUCLIDES

The ultimate fate and effects of long-lived radionuclides in the Great Lakes ecosystem are first and foremost dependent on their chemical forms which significantly influence their partitioning and chemical transport in various compartments. These parameters, in conjunction with the physical transport of water masses and associated suspended particles, play an

important role in determining the availability of the radiation to the living matter. This section of the review, therefore, summarizes our knowledge regarding these parameters.

Solution Forms of Radionuclides

The chemical forms of radionuclides in dissolved form are influenced by factors such as Eh, pH, and the composition of lakewater. The dominant solution species of most naturally-occurring radionuclides and common transuranic activation products have been predicted from thermodynamic data in the Eh-pH range of Lake Ontario waters (Platford and Joshi, 1986). Their findings are summarized in Table 1; Pu may also exist in hexavalent form and, if the solubility is exceeded, as the tetravalent oxide associated with suspended solids. No attempts as yet have been made to characterize the radionuclide species in any of the Great Lakes except Lake Michigan where dissolved $^{239,240}Pu$ is largely present in the oxidation states V and VI (Wahlgren et al., 1977; Wahlgren and Orlandini, 1982). The relative stabilities of Pu(V) and Pu(VI) are difficult to predict but evidence has been provided (Nelson and Orlandini, 1979) that the oxidized form of plutonium in Lake Michigan waters is Pu(V); it has also been shown that Pu(V) is rapidly reduced to the (IV) state in the presence of natural sediments.

Charge characteristics of plutonium in Lake Michigan water (Alberts et al., 1977) suggest that the radioelement does not associate with colloidal matter and that it is almost quantitively absorbed by anion exchange resins. Later studies (Nelson et al., 1985) have shown that, on a weight basis, both colloidal organic carbon and suspended sediment particles have similar affinities for dissolved Pu(III) or Pu(IV).

The chemical forms (and interactions) of various radionuclides may be inferred from the known behaviour of their stable analogues in the system at hand. This is a valid assumption if the two elements or isotopes have the same origins and display identical solution chemistries and fractionation behaviour. If a radioisotope is used as an analogue, it must be ensured that both radionuclides have compatible recoil behaviour. This is particularly important for alpha- and strong beta-particle emitters. In some instances, the dissolved concentrations of certain radionuclides may be considerably lower than those of stable isotopes, so their chemical behaviour will be governed any way by the stable isotopes or by the dominant member of the same subgroup in the periodic table. Such a situation prevails in Lake Ontario waters (Platford and Joshi, 1989) where the concentrations of ^{60}Co, ^{137}Cs and ^{210}Pb are several orders of magnitude lower than those of stable isotopes or members of the same subgroup of elements. However, the chemical

forms of stable analogues of several radionuclides in the Great Lakes have also not been investigated. In other cases (e.g., actinides), true stable analogues simply do not exist, yet meaningful information with respect to the interaction of Pu(IV) with Lake Michigan sediments has been derived (Nelson and Metta, 1983a) by analogy with more abundant ^{228}Th and ^{234}Th radionuclides which exist only in the (IV) oxidation state.

Ecosystem Partitioning of Radionuclides

A knowledge of the partitioning of radionuclides in an ecosystem is essential to an assessment of the potential for build-up and transport in various compartments. This information forms the basis for evaluating the significance of sinks as well as the possible slow release of radionuclides from these sinks into the biosphere where they may present hazard long after the original discharge has ceased. In this context, an aquatic system is generally considered to be comprised of three distinct reservoir compartments: water, deposited sediments and biota. Within this framework, the partitioning of ^{137}Cs in Lake Ontario (Bowen, 1974; Durham and Joshi, 1984; Joshi, 1984) is shown in Fig. 7. It is observed that ^{137}Cs, a particle-reactive radionuclide, is present in all three major compartments with the bulk being in surface sediments; significant enrichment in various biota is also noted. Such partitioning data are

available largely for ^{137}Cs in all Great Lakes except Lake Superior. Measurements for other radionuclides, particularly ^{90}Sr, are very sparse. Although biological components have been shown to accumulate radionuclides in all the Great Lakes (Yaguchi et al., 1973; Waller et al., 1973; Bowen, 1974; Joshi, 1984), precise mechanisms and factors controlling their uptake (and possible later release) have not been investigated.

Nearly all partitioning studies in the Great Lakes have focused on sediment/water interactions of the radionuclides. Only one study (Platford and Joshi, 1989) has explored radionuclide partitioning across other natural interfaces such as surface microlayer and foam accumulating in the pool just below Niagara Falls. Their findings, shown in part in Fig. 8, suggest that foam and surface microlayer are also efficient collectors of ^{60}Co, ^{137}Cs, ^{210}Pb, ^{226}Ra, and thoriums.

Although the small volume and mass of the surface microlayer preclude the possibility of its being an important reservoir of radionuclides relative to the total lake volume, its enrichment in radionuclides and other substances including microorganisms (Owen and Meyers, 1984) suggests that this zone plays an important role in the ecological cycling of radioactive substances.

Sediment/Water Interactions

Radionuclides such as ^{137}Cs and $^{239,240}Pu$ have large sediment/water distribution coefficients (Wahlgren and Nelson, 1976) and short residence times (when compared with the water retention times) in the Great Lakes (Wahlgren and Nelson, 1973; Edgington and Karttunen, 1977; Wahlgren et al., 1980). Consequently, the bulk of such radioactivity resides in the bottom sediments which thus constitute a significant radionuclide reservoir within the aquatic ecosystem. Whether sediments act as the final sink for this radioactivity or allow its remobilization to the overlying waters is largely dependent upon the nature of the biogeochemical interactions in the system.

Alberts et al. (1974) have shown that $^{239,240}Pu$ is primarily associated with the hydrous oxide coatings of Lake Michigan sediments, while ^{137}Cs is associated with the mineral fractions of these radionuclides. Americium-241 (half-life 432y), another important fallout transuranic which is continually being produced by the decay of fallout precursor ^{241}Pu (half-life 14.4y) present in sediments, is also likely associated with the hydrated oxides of iron and manganese. Thus, significant quantities of highly toxic transuranics are unlikely to reenter the water column unless the sediments become sufficiently anoxic to permit dissolution of the hydrated oxides. Such reducing conditions may

occur via the redistribution of sediment by physical transport (Edgington and Robbins, 1975) to an anoxic regime or may arise as a result of annual thermal cycling.

The potential for physical transport of sediments is indicated by the concentration-size relationships. Alberts and Muller (1979) have described the distribution of ^{137}Cs and plutonium in various particle size classes of Lake Michigan sediments. Their findings, in part, are shown in Fig. 9 from which it can be inferred that the distributions of these radionuclides are not a function of particle size. This behaviour is in sharp contrast to that expected on the basis of earlier results which show that the preferred association of ^{137}Cs and $^{239,240}Pu$ with clays (Francis and Brinkley, 1976) and hydrated oxides of iron and manganese (Alberts et al., 1974), respectively, would strongly favour partitioning of these otherwise dissimilar radionuclides in the $\leq$ 2 μm fraction.

Alberts and Muller (1979) have also shown that the sediment/water distribution coefficient is high and constant with particle size and depth so that even if sediments from few cm are resuspended, the radionuclides will remain predominantly associated with the solid particles. A subsequent study (Alberts and Orlandini, 1981) showed that very little $^{239,240}Pu$ (and ^{241}Am) are released from lake sediments under oxic and anoxic conditions and

that neither radionuclide is recycled from the sediments to the overlying water during anaerobic conditions accompanying thermal stratification. These investigators also found that the adsorption of the two radionuclides is not correlated with extractable iron or manganese. Thus, the role of hydrated oxides in the biogeochemical cycling of transuranics remains undefined.

Wahlgren et al. (1980), on the other hand, have suggested that biogenic silica and calcite mediate the seasonal cycling of $^{239,240}Pu$ in Lake Michigan (Fig. 10). They found that losses of plutonium from the epilimnion cannot be accounted for by residence time parameters. It was postulated that enhanced removal of plutonium results from intense particle production during spring and summer months and that plutonium is scavenged by silica and calcite particles which subsequently redissolve. A possible difficulty in accepting this mechanism may lie in the implication that all oxidation states of plutonium have somewhat similar behaviour insofar as their interaction with calcite and silica is concerned.

The strong affinity of atmospherically-delivered ^{210}Pb for suspended particulate matter in the Great Lakes is well known (Durham and Joshi, 1980a; Eadie and Robbins, 1987; Van Hoof and Andren, 1989) and forms the basis for dating bottom sediments (Robbins and Edgington, 1975a; Farmer, 1978; Nriagu et al., 1979;

Durham and Joshi, 1980a; Christensen and Chien, 1981; Evans et al., 1981; Joshi, 1985). Recent investigations (Eadie and Robbins, 1987; Van Hoof and Andren, 1989) report a strong affinity of ^{210}Pb with calcite. Van Hoof and Andren (1989) also noted that the season-dependent partitioning of this radionuclide in Lake Michigan is fairly constant with depth except during calcite precipitation in the epilimnion and for larger particles in the nepheloid layer; zooplankton and newly formed diatoms play a minor role when compared with that of calcite. In all, therefore, calcite precipitation appears to play a major role in modulating the seasonal variation of radionuclides of diverse origins.

Other possible mechanisms whereby radionuclides from deposited sediments may be released to the overlying waters include sediment reworking by deposit-feeding organisms (Robbins et al., 1977; Robbins, 1982) and diffusion from pore water (Lerman and Taniguchi, 1972; Tracy and Prantl, 1983). Diffusion of a radionuclide from pore water to the overlying water will occur only if a concentration gradient is established. No direct measurements on the pore water concentration of any radionuclide have been reported as yet in the Great Lakes system. In absence of such information, Tracy and Prantl (1983) have estimated the concentration, C_{iw}, of ^{137}Cs in the interstitial water of Lake Superior sediments by using the relationship

$$C_{iw} = C_s \times \rho \times (V_s/V_w) \times (1/K_d) \times 1000 \text{ cm}^3 \text{ L}^{-1} , \qquad (2)$$

where the concentration of ^{137}Cs in sediment, C_s, is 1.11 Bq g^{-1} dry weight, the density of dry sediment, ρ , is 2.5 g cm^{-3}, the ratio of sediment volume to water volume, V_s/V_w, is 0.15/0.85, and K_d is the ^{137}Cs equilibrium distribution coefficient (dimensionless) between sediment and water. Setting K_d at 4000 (Robbins et al., 1977), Tracy and Prantl (1983) calculate a value of 122 mBq L^{-1} for C_{iw}. Since this value is significantly higher than that prevailing in the open waters of Lake Superior, Tracy and Prantl (1983) suggest that ^{137}Cs is reentering water from sediment.

The value of K_d employed by Tracy and Prantl (1983) was obtained by Robbins et al. (1977) for the surface sediments of Lake Huron. It is not clear whether this value was derived from measurements on sediment porewater or lakewater or how it was inferred that this value represents equilibrium value. Wahlgren and Nelson (1976) have reported a sediment/lakewater K_d value of 3×10^5 for ^{137}Cs from measurements on 8 samples from Lake Michigan. This value is very similar to that inferred for the lower Great Lakes (Joshi, 1988b). The available data (Durham and Joshi, 1984) show that ^{137}Cs concentrations in the Great Lakes waters showed little year-to-year variation from about 1975 to 1981, thus approaching a near equilibrium state. Setting K_d at 3×10^5 in equation (2), a value of 1.6 mBq L^{-1} is obtained for

C_{iw}. This estimate is very similar to the average concentration of about 1.8 mBq L^{-1} measured (Durham and Joshi, 1984) in the open waters of Lake Superior during the same time period. It therefore appears more reasonable that, contrary to the proposal by Tracy and Prantl (1983), the diffusional migration of porewater ^{137}Cs to lakewater is unlikely to occur. Alberts et al. (1989) also found no evidence for the diffusional release of ^{137}Cs from the sediments to the overlying waters in Lake Michigan where this radionuclide is almost irreversibly bound to the mineral fraction of the sediments.

DISTRIBUTION OF RADIONUCLIDES IN THE GREAT LAKES

Lake Water

Pursuant to the 1972 U.S./Canada Great Lakes Water Quality Agreement (GLWQA, 1972), systematic monitoring of the Great Lakes open waters for radionuclides started in 1973. Samples collected in 1973 were analyzed for ^{90}Sr and $^{239,240}Pu$ by the Argonne National Laboratory (ANL) while concentrations of ^{3}H and all gamma-emitting radionuclides were measured at the National Water Research Institute (NWRI). Since this preliminary assessment, ANL continued to analyze water samples for plutoniums (Nelson and

Metta, 1983b) while NWRI continued the analyses for the remaining radionuclides (Durham and Joshi, 1984) until 1982 when another assessment of data was made. In view of the prevailing very low levels, it was recommended (IJC, 1989) that synoptic open water surveys be made every 5-10 years. Environment Canada plans to conduct first such survey in 1990-91.

The results for samples collected in 1973 and 1981 are given in Fig. 11. These values indicate that the levels for most radionuclides declined at a rate of 2-5% per year. The only exception is tritium in Lake Ontario which increase is attributable to the enhanced releases of this radionuclide due to the expansion of nuclear power generating capacity along its shores since 1973. Available data (Baweja et al., 1987) on the levels of tritium in Niagara River and St. Lawrence River shows that the 1982-84 levels of this radionuclide in the two rivers were below those measured in the open waters thereby suggesting that inputs from nuclear facilities along the shores of Lake Ontario are considerably diluted before exiting the system.

Assuming the lake waters are well mixed and that the fallout flux of chemically-conservative ^{90}Sr to the lakes has been constant (i.e., ignoring the difference in fallout rates to upper and lower Great Lakes indicated in Appendix 1), the relative levels of this radionuclide in these lakes should be

inversely proportional to their mean depths. The only exception is Lake Erie where the residence time of water, 2.6 y (CCGLBHHD, 1977), is short compared to the time since the major fallout occurred. This trend is well reflected in the 1973 measurements (Fig. 11) when contributions from nuclear facilities or land runoff were much lower. Thus the level of ^{90}Sr in Lake Superior is about half those of Lakes Michigan, Huron, and Ontario, which follows from its mean depth at 147m (CCGLBHHD, 1977) being about twice the mean depths of the other lakes, 85, 59 and 86m, respectively. The levels of particle-reactive ^{137}Cs and $^{239,240}Pu$ in water, on the other hand, are controlled by the sedimentation rates. Lake Erie with the highest sedimentation rate in the system, therefore, has the lowest levels of these radionuclides.

Besides radionuclides listed in Fig. 11, relatively short-lived radionuclides such as ^{125}Sb (half-life 2.8y) and ^{144}Ce (half-life 284d) have also been detected in the open waters of the Great Lakes (Durham and Joshi, 1984). ^{144}Ce was detectable in Lakes Superior and Huron in 1974 and 1975, while ^{125}Sb was present in measureable amounts until about 1982-83. Measurements on the levels of ^{241}Am are very scant though the levels in Lake Michigan (0.5 $\mu Bq\ L^{-1}$; Orlandini, 1979) are comparable to those of $^{239,240}Pu$. Occurrence of fallout ^{113m}Cd (half-life 14.6y) in Lake Michigan at extremely low levels (350 $\mu Bq\ L^{-1}$) has been reported recently (Dunn and Tisue, 1989).

Only few data exist on the levels of naturally-occurring radionuclides in the Great Lakes ambient waters. Durham and Joshi (1980a) reported a concentration of about 0.7 mBq L^{-1} for ^{226}Ra in Lake Huron waters while ^{210}Pb was present at a level of about 0.5 mBq L^{-1}. Uranium and ^{226}Ra levels in Lake Ontario average about 0.7 µg L^{-1} and 1.2 mBq L^{-1}, respectively (Baweja et al., 1987). Levels of ^{228}Th in Lake Michigan average about 28 µBq L^{-1} (Nelson and Metta, 1983a).

Site-specific monitoring of nearshore waters for various radionuclides is performed by numerous other Canadian and U.S. federal, provincial or state agencies (IJC, 1979 and 1983) as well as the industry (Ontario Hydro, 1987). In general, the levels reported todate are similar to those measured in the open waters. In Canada, the Department of National Health and Welfare has maintained a drinking water monitoring program since 1963 (IJC, 1983). Current emphasis of this program appears to be on naturally-occurring radionuclides (HWC, 1987), although unplanned releases of reactor tritium have also been monitored via measurements on drinking water (IJC, 1989).

Results from radionuclide measurements on floc samples from drinking water filtration plants located near the Pickering nuclear generating station on the shores of Lake Ontario indicate (Durham and Joshi, 1981) that many particle-reactive

radionuclides are retained to some degree on the floc samples. Available measurements (S.R. Joshi and R.C. McCrea, unpublished data) on raw and filtered waters and floc samples from a water filtration plant near Ottawa show that radiologically-important, conservative radionuclides such as ^{90}Sr are not retained on the floc. Results from measurements on the raw and drinking water samples from Lake Michigan (Alberts and Wahlgren, 1977) show that varying amounts of $^{239,240}Pu$ will also be retained on the floc.

Sediments

The surficial sediments of the Great Lakes, with the exception of southern Lake Michigan (Cahill and Steele, 1986), have not been systematically surveyed for the levels and distribution of any of the radionuclides. Much of the information on anthropogenic radionuclides in Great Lakes sediments is the result of vertical profile measurements intended to derive estimates of sedimentation rates (Karttunen and Edgington, 1974; Robbins and Edgington, 1975a; Edgington and Karttunen, 1977; Durham and Joshi, 1980a; Christensen and Chien, 1981; Joshi, 1985). Few investigations have focused on the behaviour of these radionuclides as pollutants (Bowen, 1974; Joshi, 1988b and 1988c; Rodgers and McKinley, 1988).

The distribution of ^{137}Cs and $^{239,240}Pu$ in the Great Lakes sediments is shown in Figs. 12 and 13, respectively. The patterns shown pertain to total amounts of these radionuclides as opposed to commonly described pollutant distributions in the upper few cm of sediments which method underestimates the inventories available for possible release or relocation.

Radionuclide inventory estimates for Lakes Michigan, Erie and western Lake Ontario were directly taken from literature (Edgington and Karttunen, 1974; Karttunen and Edgington, 1977; Joshi, 1988b and 1988c), while those for Lakes Huron and other depositional basins in Lake Ontario were inferred from separately reported analytical data and sedimentation rate values (Bowen, 1974; Farmer, 1978; Durham and Joshi, 1980a; Joshi 1985 and unpublished data). The ^{137}Cs inventories in Lake Superior are based on unpublished NWRI measurements and reported (Evans et al., 1981) sedimentation rates. The distributions depicted in Figs. 12 and 13 represent recent (1989) estimates as inferred using fallout inputs (Appendix 1) since measurements were first made and sedimentation rates; the decay of ^{137}Cs was also taken into account. Because of the limited number of sediment cores analyzed todate, these distribution patterns involve significant extrapolation of data and should be considered as highly approximate.

The radionuclide deposition patterns shown in Figs. 12 and 13 largely reflect fallout inputs to major depositional basins as most measurements pertain to the time period before significant nuclear power production commenced in the basin. The only exception is western Lake Ontario which is known (Joshi, 1988b and 1988c) to be impacted by releases from a nuclear facility. Generally higher concentrations of the two radionuclides are observed near river (or connecting channel) mouth locations where additional fallout activity may be supplied from the watershed. Robbins and Edgington (1975b) have suggested that enhanced radionuclide scavenging by river-borne sediment may account for elevated radionuclide levels near river mouths. Nelson et al. (1984) however have shown that river inputs to Lake Michigan in fact reduce the overall lake concentrations via dilution with less active materials.

Alternatively, it may be suggested that resuspension and interlake translocation of deposited sediment result in elevated radionuclide levels at connecting channel mouths. Edgington and Karttunen (1977) have shown that interbasin migration of ^{137}Cs can occur in Lake Erie sediments. These investigators also found that the Lake Erie sediments account for about 75% of the expected inventory. The remaining activity is then obviously carried to Lake Ontario via the Niagara River which has negligible radionuclide and sediment contributions from its

drainage basin. Thus the observed higher radionuclide levels and sedimentation rates at the Niagara River mouth locations in Lake Ontario (Joshi, 1988b and 1988c) arise in part due to contributions from Lake Erie.

Biota

Few data exist on the levels of radionuclides in the Great Lakes biota. Most of the existing measurements were made in the early 1970s (Waller et al., 1973; Yaguchi et al., 1973; Bowen, 1974). Since then only occasional measurements have been reported on open lake fish samples (Joshi, 1984) though fish from the vicinities of nuclear power plants have been analyzed on regular basis by the industry (see, for example, Ontario Hydro, 1987 and other reports in the series.)

Results from the above-cited measurements on ^{137}Cs in biota from the five lakes are shown in Fig. 14. The lakewater concentration factors were calculated by dividing the measured activity of ^{137}Cs in organism (fresh weight) by that in water. The results indicate increasing concentration factors for this radionuclide for more complex organisms. This reconcentration mechanism is of concern since the ultimate predator at the highest level is man. However some radionuclides may not display a similar 'trophic level' effect. For example, in the case of

$^{239,240}Pu$, piscivorous fish have the lowest concentration factors (Fig. 15). Marshall et al. (1975) have concluded that the reduction in plutonium levels along the food chain reflects the settling of phytodetritus or zooplankton fecal pellets as these two organisms adsorb much more plutonium than ^{137}Cs from water in the first place. The precise mechanisms of the different behaviour of the two radionuclides in the subsequent steps in the food web are not known.

The 'trophic level' behaviour of most other radionuclides in the Great Lakes aquatic food chains has not been examined; the limited available data (Joshi, 1984) suggest that uranium is not accumulated along the food chain while ^{226}Ra shows some accumulation in the piscivorous fish. While not unequivocally established, there is some evidence (Joshi, 1984) that the levels of ^{137}Cs and ^{226}Ra may be declining.

Besides being a source of radiation to the humans, radioactivity in the fish also provides radiation dose to the species themselves (Joshi, 1984; Environment Canada, 1986). This radiation dose may be organ-dependent as the radionuclide may be inhomogeneously distributed in fish (Platford and Joshi, 1988). Information on the distribution of radionuclides in various organs of the Great Lakes fish is sparse. The only available data (Bowen, 1974) are shown in Fig. 16 from where it can be

inferred that both ^{137}Cs and $^{239,240}Pu$ have dissimilar distribution in various species.

INTERLAKE TRANSPORT OF RADIONUCLIDES

The movement of water and sediment phases plays considerable role in the overall distribution of radioactivity in the Great Lakes. Two studies have described the movement of tritium - a radionuclide unlikely to show preferential association with sediment - released to the lakes. The first study (Lam and Durham, 1984) explained the migration of a controlled release of tritium from the Pickering nuclear power generating station. A simple transport and diffusion model using empirical length-scale-dependent eddy diffusivities produced satisfactory results in simulating a moving patch of tritium in Lake Ontario.

The second study (Veska and Tracy, 1986) assessed the transport of two exceptional releases of tritium from the Bruce and (the then operating) Douglas Point nuclear power generating stations. A straightforward application of the classical diffusion equation adequately described the travel times between the point of release and the sampling location in Lake Huron.

The long-term behaviour of radionuclides in the Great Lakes waters is also strongly influenced by hydrologic parameters. Lerman (1972) has described a physical model for the chain of well-mixed Great Lakes in which outflows from both Lake Superior and Lake Michigan feed into Lake Huron which discharges into Lake Erie. The outflow from Lake Erie feeds into Lake Ontario, the last lake in the system. The concentration-time model was used to describe the transport of fallout ^{90}Sr, a radionuclide which is predominantly present in the solution form (Lerman and Taniguchi, 1972). The input function for each lake considered contributions from direct fallout over the lake water surface, water flow from land and inflow from the preceding lake(s) as applicable, while removal was by outflow and decay only. Lerman (1972) evaluated the model for assumed contributions of up to 10% of annual deposit in the land drainage basin. His results assuming no contribution from the drainage basin are shown in Fig. 17. A better agreement between computed and observed concentrations was obtained if both the ^{90}Sr input and rates of removal were assumed to be higher. According to this model, seasonal stratification of the lakes does not affect the mean annual concentrations of this radionuclide.

The results shown in Fig. 17 also indicate that lower Great Lakes are influenced by the upstream lakes. This influence is particularly noticeable in the case of Lake Ontario where the

peak concentration lags by at least 1 y. Also, though all the lakes have been assumed to receive an equal flux of this radionuclide, their responses differ significantly due to different mean depths of the lakes. Both these hydrologic effects have been illustrated by Chapra and Reckhow (1983) using a different computational framework and an impulse function to idealize the continuous loading of ^{90}Sr. Their results are also given in Fig. 17. The simulation, however, cannot be compared with that given by Lerman (1972) since Chapra and Reckhow (1983) use a different input function and ignore the drainage basin contribution.

Unlike ^{90}Sr, fallout $^{239,240}Pu$ has strong affinity for sediments and soils and is a much longer-lived radionuclide. Consequently, drainage basin contributions and radioactive decay can be ignored, but losses due to sedimentation must be considered. Wahlgren et al. (1980) have evaluated the behaviour of fallout plutonium under these conditions using a model otherwise equivalent to that given by Lerman (1972). Their results are also shown in Fig. 17. Clearly, the model satisfactorily accounts for the observed concentrations of this radionuclide in the Great Lakes waters. Upon closer inspection of the data, Wahlgren et al. (1980) found that an even better fit to measurements for Lake Michigan can be obtained if it is assumed that 0.05% of the drainage basin inventory of this radionuclide

is lost to the lake. Like ^{90}Sr, little is presently known about the drainage basin contributions of this radionuclide. It also remains to be seen what influence such an assumption will have on the predicted concentrations in the lower Great Lakes where the drainage basin/lake surface area ratio is about 50% higher than that for Lake Michigan (CCGLBHHD, 1977).

Model calculations by Wahlgren et al. (1980) yielded mean residence times of around 2-3y for plutonium in all the lakes except Lake Erie where a value of 0.4y was obtained. Their results clearly show that this residence time is significantly controlled by both the outflow of water to the next lake and transfer to the bottom sediments on settling particles.

Tracy and Prantl (1983), on the other hand, report that the persistence of ^{137}Cs, a radionuclide with affinity similar to that of $^{239,240}Pu$ for sediments, in Lake Huron and Lake Superior waters is best explained in terms of a two-component residence time with the short-lived component (0.3-0.7y) accounting for removal by settling. However, Tracy and Prantl (1983) have completely ignored the inflow/outflow characteristics of the two lakes whose role in regulating radionuclide concentrations in Great Lakes waters is well established from other investigations (Lerman, 1972; Chapra and Reckhow, 1983).

In all, it appears that Lerman's (1972) original model and variation thereof (Wahlgren et al., 1980) can adequately predict the long-term behaviour of both conservative and particle-reactive fallout radionuclides in the Great Lakes waters. However, a further validation of these models is warranted in view of the revised estimates of fallout inputs (Appendix 1) and the possible future availability of reliable estimates of radionuclide contributions due to watershed runoff.

RADIOLOGICALLY DEGRADED AREAS IN THE GREAT LAKES BASIN

Serpent River, Lake Huron

The Serpent River watershed drains the Elliot Lake uranium mining area and discharges into the Serpent Harbour on the north shore of Lake Huron. The watershed has been receiving enhanced radionuclide loadings since about 1955. The Serpent River waters have been regularly monitored by the Ontario Ministries of Environment (MOE) and Labour (MOL) for various radionuclides. Their results showed that mining activities have led to some impairment of the radiological water quality in the river basin with respect to ^{226}Ra. Implementation of remedial measures began in 1966. Since then the concentrations of ^{226}Ra in the river

(Fig. 18) have shown gradual decline, and by 1985 all water quality criteria for this radionuclide had been met.

The levels of radionuclides (and of some metals associated with the uranium ore) in the Serpent Harbour sediments were first measured in 1975 in a joint study (Ross and Chatterjee, 1977) by MOE and the Canada/U.S. International Joint Commission (IJC). A follow-up Environment Canada-sponsored investigation (Hart and McKee, 1985) found that the 1984 concentrations of uranium in the sediments represented about 40% of their 1975 levels. This study also found that the concentrations of several trace metals in the sediments exceeded provincial guidelines for the open water disposal of dredged spoils. Such guidelines with respect to radioactive contaminants have not been developed as yet. The Serpent River watershed is continually monitored by the provincial agencies.

Cattaraugus Creek, Lake Erie

Cattaraugus Creek receives effluents from the Western New York Nuclear Service Center (WNYNSC) located at West Valley, NY, about 65 km upstream of Lake Erie. The facility comprises the first commercial nuclear fuel reprocessing plant in the United States and various storage, treatment and burial areas for radioactive wastes. Although no spent fuel has been reprocessed

since 1972, releases of controlled amounts of radioactivity to the local watershed have continued. The average 1969-71 ^{90}Sr levels resulting from such releases to Cattaraugus Creek failed to meet both the U.S. Environmental Protection Agency's standard for drinking water and the U.S. Nuclear Regulatory Commission's technical specifications for the creek (IJC, 1983). Since about 1973, levels of this radionuclide in the creek (Fig. 19) have continually met both the above criteria.

Ecker and Onishi (1979) and Onishi et al. (1981) have extensively studied the local aquatic system. Their results show that Cattaraugus Creek sediments and waters have been impacted by releases from WNYNSC and that this radioactivity is detectable at a station near the confluence of the creek and Lake Erie. Joshi (1988b, 1988c) has demonstrated that West Valley-derived radionuclides are subsequently transported to Lake Ontario via the Niagara River. Most of the particle-reactive radionuclides reside in the bottom sediments of Lake Ontario. Results for $^{239,240}Pu$ (Joshi, 1988c) are shown in Fig. 20. The prevailing high sedimentation rates in the area allow a clear distinction to be made between the 1963 fallout peak and the 1969-71 discharge from the WNYNSC site. These results unambiguously establish that any accidental releases from WNYNSC will be transported to Lakes Erie and Ontario where they will provide additional radiation dose to area residents through drinking water supplies.

The aquatic system in the vicinity of WNYNSC has been continually monitored by the New York State Department of Environmental Conservation (NYSDEC, 1967-81) and by the Department of Health (1982-88) for a variety of radionuclides. The low-level waste burial area does not accept any new radioactive wastes. The former nuclear fuel reprocessing facility is now being used to investigate methods of encapsulation of high-level waste currently stored on site.

Port Hope, Lake Ontario

Port Hope Harbour was the site of a radium recovery plant from 1933 to 1952 and, since 1955, has been the site of a uranium refinery. The refinery included a UO_2/UO_3 production facility, which ceased operation in 1983, and a UF_6 production facility which has operated since 1970. The UF_6 production was expanded in 1984 with the addition of a second plant. As a result of waste discharges, concentrations of uranium and gross α and gross β radioactivity in the harbour water are often above maximum acceptable values (IJC, 1989). In addition, about 90,000 m^3 of sediments located in the turning basin and west slip harbour areas are contaminated with uranium- and thorium-series radionuclides, heavy metals and PCBs. Contamination is believed to be primarily the result of waste management practices associated with refining operations although urban runoff and

other industrial sources have undoubtedly impacted the harbour. The harbour has been designated an Area of Concern by the IJC's Great Lakes Water Quality Board on the basis of sediment contamination. Due to their radioactivity content, the harbour sediments have been classified as a low-level radioactive waste.

Detailed surveys of the Port Hope Harbour sediments were carried out by MOE and MOL in 1968 and 1971; a third survey was jointly carried out by MOE and Environment Canada in 1978 (Ogilvie, 1981; Durham, 1981). Based on these measurements, Durham (1981) concluded that the ^{226}Ra contamination of the harbour sediments has very little effect on the human food chain though potential risk to human health exists from direct contact with the sediments. A subsequent Environment Canada-sponsored study (McKee et al., 1985) basically confirmed earlier findings and provided some additional data on the distribution of both radioactive and non-radioactive contaminants in sediments and benthos. This investigation found that nitrogen and heavy metal concentrations at some locations in the harbour exceeded provincial guidelines for open water disposal of dredged spoils.

In the 1968 survey of the harbour, Cook and Veal (1968) noted that most of the chironomids in the inner harbour had deformed head capsules. Warwick et al. (1987) have quantified the incidence of deformities in differentially polluted areas of

the harbour. Their findings suggest that though radiation dose may be a significant factor in the induction of deformities, heavy metals and elevated water temperatures may also be involved. Another Environment Canada investigation (Environment Canada, 1986) has concluded that the prevailing radionuclide levels in the area fish do not constitute significant hazard to the fish or to the population consuming these fish. Uranium levels in airborne emissions from the refinery (Tracy and Meyerhof, 1987) or those associated with the application of Port Hope sewage sludge on agricultural lands (McKee and Lush, 1987) are also unlikely to constitute any observable health effects.

Environment Canada and the MOE have developed a Remedial Action Plan (RAP) for Port Hope Harbour. The RAP will require a proper dredging protocol and confined disposal of dredged materials and as such its implementation is largely dependent upon the establishment of a low-level radioactive waste disposal facility. Meanwhile, in addition to continuing monitoring programs by MOE and others, two other studies are underway. In the first study, Atomic Energy of Canada Limited will evaluate potential environmental risks involved in the cleanup. The second study, by NWRI, is designed to assess the potential for recontamination of harbour following cleanup.

Port Granby, Lake Ontario

The Port Granby radioactive waste management site is located about 13 km west of Port Hope. The shorline in the region consists of steep bluffs. The waste is buried at the top of these bluffs. The main disposal area is a rectangular block extending for a distance of about 300m along Lake Ontario. Open dumping of wastes from the Port Hope refinery commenced in 1955. Subsequently, this disposal technique was changed to trench containment, although some liquid waste was discharged to the ground from 1974-76. The waste at Port Granby is primarily comprised of ^{230}Th and its decay products, notably ^{226}Ra, traces of uranium, and significant quantities of non-radioactive constituents such as arsenic and nitrate. Two creeks drain the site and run into artificial catchbasins located about 50m from the lakeshore.

The concentrations of ^{226}Ra, arsenic and nitrate in Lake Ontario waters off Port Granby were first measured in 1977 (Durham and Joshi, 1980b). These measurements showed that the leachate plume, moving parallel to the shoreline in the direction of the prevailing wind, has only a minor effect on the lakewater quality. Subsequently, in order to study the migration of contaminants to the groundwater, rows of piezometers were

installed in 1981-83. The results on groundwater measurements showed (Platford et al., 1984) that about 2.5 x 10^5 m^3 of groundwater flowing annually from the site into Lake Ontario carries with it about 25 MBq of ^{226}Ra and 25 kg of uranium. Both of these contaminants however are diluted, within a metre or so from the shore, to meet the Canadian drinking water guidelines.

The groundwater contaminant transport at the site has been thoroughly evaluated and mathematical models developed and verified (Bobba and Joshi, 1988 and 1989) using field data. The predictive modelling findings for ^{226}Ra are summarized in Fig. 21 in terms of possible future relocation of waste to another site. The model predicts that even if all the waste is removed from the site, this radionuclide will continue its slow migration, via groundwater, toward Lake Ontario and it will take over 1000y before significant decline in levels at the lakeshore locations is observed. The levels at the current waste site will record a faster, gradual decline. Decommissioning plans for the site have been prepared as erosion of the bluffs threatens to expose the wastes within the next few decades. New site however has not been selected as yet. Meanwhile, investigations into bluff stability are continuing at NWRI.

STANDARDS, CRITERIA AND OBJECTIVES

The impact of radioactive contaminants present in the Great Lakes has traditionally been assessed in relation to human health. Little consideration has been given to the potential impact of this contamination on the ecosystem health. As discussed earlier in the text, several aquatic organisms tend to accumulate radionuclides. Only few estimates (Joshi, 1984; Environment Canada, 1986; Platford and Joshi, 1989) are available for the radiation dose absorbed by these organisms; the ecosystem significance of this chronic, low-level irradiation has not been studied. Thus the effect of this radiation exposure needs to be evaluated and objectives developed. The need is particularly obvious for fish the consumption of which also constitutes a pathway for radiation dose to humans as will be seen later in the text.

Radiation dose to humans in the Great Lakes basin has usually been inferred from concentrations of selected radionuclides in lake waters. Several U.S. and Canadian jurisdictions have established limitations for the concentrations of radionuclides in drinking water (IJC, 1983). The objective for radioactivity in the Great Lakes (IJC, 1979 and 1983) is also

based on the consumption of drinking water and requires that the dose equivalent commitment to an individual drinking the lake water (outside of any defined source control area) should not exceed 10 μSv per year. Since all these standards or objectives are based on different assumptions (IJC, 1983), dissimilar limits may be obtained for the concentration of the same radionuclide as is shown in Figs. 18 and 19. A discussion of the origins and significance of these differences is beyond the scope of the present review. The need for arriving at a uniform standard is, however, quite obvious. The IJC water quality objective for radioactivity nearly meets this need as it was jointly developed by the U.S. and Canadian agencies.

As noted above, the radioactivity objective is based solely on the consumption of lakewater. Food (primarily fish) harvested from the Great Lakes and consumed by man is another pathway of radionuclides to man. This pathway has been considered insignificant in previous assessments (IJC, 1979). The evidence to the contrary, however, is beginning to emerge. Table 2 gives estimates of ^{137}Cs radiation dose received due to the consumption of Great Lakes fish. These estimates were obtained using the reported levels (Joshi, 1984) and the dose conversion factor (IJC, 1983) for this radionuclide; it was assumed that the edible portions of the fish retain 50% of the radioactivity. The actual dose received will ofcourse depend upon many other factors

including consumption patterns. This preliminary calculation shows that, for this radionuclide, the consumption of 1 kg (about 5 servings) of fish imparts about two times the radiation dose (0.02 μSv; see later in the text) resulting from the annual consumption (803 L) of average lakewater.

A recent study (Environment Canada, 1986) assessed the radiation doses due to the consumption of Port Hope Harbour fish containing six naturally-occurring radionuclides (^{210}Pb, ^{226}Ra, ^{228}Th, ^{230}Th, ^{232}Th, and U.) The results for the edible portion of rainbow trout collected from Ganaraska River area are shown in Table 3. Previous measurements (Joshi, 1984) have shown that the levels of ^{226}Ra and U in the area fish have substantially declined and are virtually indistinguishable from those measured in fish from other Great Lakes. The doses were inferred using three different sets of concentration-to-dose conversion factors (CDCFs). The Johnson and Dunford (1983) CDCFs are very similar in magnitude to those suggested by the International Commission on Radiological Protection (1979) for all radionuclides except thoriums where the two methodologies assume different gut-to-blood transfer coefficients. The values of the Johnson (1986) CDCFs for thoriums range between those provided by the other two models. The most conservative of the three estimates given in Table 3 implies that an annual consumption of 1 kg of fish would result in a dose commitment similar to that (2 μSv; see later in

the text) resulting from the consumption of lakewater (containing both naturally-occurring and artificially-produced radionuclides) over the same time period.

The above dose estimates are well within the annual limit of 5 mSv recommended by the ICRP for the protection of the general public. The calculations, however, do underscore the need for an evaluation of this pathway to radiation exposure. In particular, the doses due to natural radionuclides to critical groups need special attention.

ASSESSMENT OF MONITORING DATA

The Great Lakes Water Quality Board (GLWQB) has made periodic assessments of the radionuclide monitoring data including that pertaining to atmospheric emissions (IJC, 1977, 1978, 1979, 1983 and 1989). Durham and Joshi (1984) assessed the radiation dose commitments from the 1973-81 levels of artificially-produced radionuclides in the open waters of the Great Lakes. These assessments concluded that radiation doses due to anthropogenic radionuclides in lakewater are well below the objective dose.

The concentration of uranium in water from Port Hope Harbour however continues to exceed the Canadian drinking water guideline of 20 μg L^{-1} (IJC, 1989). Recent estimates (Baweja et al., 1987; S.R. Joshi, unpublished data) of radiation dose commitment arising from the consumption of Lake Ontario ambient waters are given in Table 4. Taken together, the five radionuclides provide about 22% of the annual dose commitment. Estimates of doses arising from other naturally-occurring radionuclides such as ^{7}Be, ^{40}K and remaining members of the natural series as well as those arising from ^{14}C are not available. Their contributions will undoubtedly result in an upward revision of the current dose estimates though the annual dose commitment will be likely within the stipulated objective.

STATUS OF MONITORING ACTIVITIES

The 1978 GLWQB assessment of monitoring data (IJC, 1979) led to the following conclusion:

> Present radioactivity surveillance activities and the data they generate are generally adequate to determine compliance with the radioactivity objective and to determine trends in the radiological quality of the

> water. The programs are, however, not adequate to determine total intake of radionuclides by man from drinking water and eating fish from the lakes, nor are the present programs adequate to determine the dispersion and fate of radionuclides in the biota and the sediment. Radioactivity surveillance activities are expected to improve in the next few years as the radioactivity surveillance plan is implemented and as drinking water monitoring requirements are strengthened.

Indeed, some improvement in surveillance activities was noticeable in the subsequent report (IJC, 1983). Since about that time, however, the Great Lakes radioactivity surveillance programmes in both the U.S. and Canada have undergone substantial cuts. Open waters have not been monitored since 1983. Similarly, radionuclide levels in open lake fish have not been measured. Source control area monitoring has continued for compliance purposes. Drinking water monitoring is also largely confined to source control areas or the vicinities. In Canada, extra resources were primarily directed toward assessments of degraded areas though open waters of Lake Ontario will likely be monitored during 1990-91. Few ongoing U.S. and Canadian research studies are largely directed toward sediment/water interactions of radionuclides in Lakes Michigan and Ontario, respectively, and

are the only sources of data for open lake environment. In all, with few exceptions, the 1978 conclusion is still valid.

FUTURE CONSIDERATIONS

This review demonstrates that while most aspects of radioactivity in the Great Lakes have been addressed to varying degrees, a true understanding and mitigation of the impact will require further multi-directional efforts along the following lines:

Research

1. Determine radionuclide trends in biota with respect to species, age and food chain variability; investigate mechanisms of radionuclide uptake; and develop methodologies to assess radiation dose to biota.

2. Investigate the behaviour of ^{99}Tc (half-life 2.1 x 10^5 y) in each lake. This is the only major

nuclear fission product (with well-known availability to biota) for which no data have been obtained for the Great Lakes.

3. Determine loading, by source and route, of ^{3}H to each lake, particularly Lakes Ontario and Huron, and determine its dispersal patterns.

4. Investigate the significance of nuclear reactor emissions of ^{14}C (half-life 5430y), ^{63}Ni (half-life 100y), and ^{32}P (half-life 14d).

5. Estimate drainage basin contributions of major fallout radionuclides and refine Lerman's interlake transport model.

6. Investigate the roles of sediment resuspension and particle and sediment/porewater interactions in the cycling of radionuclides.

Monitoring

1. Continue monitoring of lake waters for both dose assessment and transport model verification

purposes and include major natural series radionuclides.

2. Develop fish monitoring programme since fish consumption is a significant pathway of radiation dose to consumers.

3. Upgrade drinking water monitoring programme by including more sampling locations and by ensuring that both major naturally-occurring and artificially-produced radionuclides are monitored.

Objectives and Guidelines

1. Develop objectives for aquatic ecosystem components (other than water) due to the role they play in the accumulation and cycling of radioactivity. The need for a radiological objective for fish is well established.

2. Objective compliance evaluation should consider doses due to major natural series radionuclides.

3. Revise guidelines for the open water disposal of

dredged spoils to include radioactive substances.

4. The size of the source control area in the water quality objective for radioactivity was arbitrarily defined as having a 1-km boundary around the discharge outfall. The objective may need revision in this respect as the extent of this area is largely governed by the dispersal characteristics of the receiving waters.

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TABLE 1

PREDOMINANT SOLUTION SPECIES OF MAJOR NATURAL SERIES RADIONUCLIDES AND SELECTED TRANSURANICS IN LAKE ONTARIO WATERS

Radionuclide	Species
^{238}U	$UO_2(CO_3)_3^{4-}$
^{232}Th	ThO_2
^{226}Ra	Ra^{2+}
^{210}Pb	Pb^{2+}
^{210}Bi	$Bi(OH)_2^+$
^{210}Po	H_2PoO_3
^{237}Np	NpO_2^+
$^{239,240}Pu$	PuO_2^+
^{241}Am	Am^{3+}

TABLE 2

ESTIMATED ^{137}Cs RADIATION DOSE RESULTING FROM THE CONSUMPTION OF GREAT LAKES FISH

Lake	Dose ($\mu Sv\ kg^{-1}$)
Lake Superior	0.07
Lake Huron	0.06
Lake Erie	0.01
Lake Ontario	0.01

TABLE 3

NATURAL RADIATION DOSE RESULTING FROM THE CONSUMPTION OF GANARASKA RIVER RAINBOW TROUT

Model	Dose (mSv kg^{-1})
ICRP (1979)	0.002
Johnson and Dunford (1983)	0.196
Johnson (1986)	0.033

TABLE 4

ANNUAL RADIOLOGICAL DOSE COMMITMENT FROM LAKE ONTARIO WATERS

Radionuclide	Dose equivalent (μSv)
^{3}H	0.1
^{90}Sr	1.0
^{137}Cs	0.02
^{226}Ra	0.4
^{238}U	0.7

APPENDIX 1

ESTIMATED FALLOUT OF ^{90}Sr AND ^{137}Cs OVER GREAT LAKES

Year	^{90}Sr		^{137}Cs	
	Flux (MBq km^{-2} y^{-1})	Cumulative (MBq km^{-2})	Flux (MBq km^{-2} y^{-1})	Cumulative (MBq km^{-2})
		Lake Superior		
1954	62.5	62.5	100.0	100.0
1955	80.8	141.8	129.3	227.0
1956	100.2	238.6	160.3	382.1
1957	100.5	333.5	160.8	534.2
1958	139.4	465.0	223.1	745.1
1959	196.3	650.3	314.0	1042.2
1960	35.5	670.4	56.8	1075.4
1961	55.0	709.6	87.9	1138.9
1962	279.1	971.9	446.5	1559.5
1963	538.5	1487.5	861.7	2385.8
1964	358.8	1811.1	574.1	2905.6
1965	125.1	1893.5	200.2	3039.8
1966	54.8	1903.6	87.7	3058.5
1967	37.2	1895.8	59.6	3048.6
1968	29.8	1880.9	47.7	3027.1
1969	32.4	1868.8	51.8	3010.1
1970	33.4	1858.1	53.5	2995.3
1971	32.0	1846.3	51.2	2978.5
1972	17.0	1819.7	27.2	2938.0
1973	9.4	1786.1	15.1	2886.3
1974	21.1	1765.0	33.7	2854.5
1975	15.4	1738.7	24.6	2814.3
1976	3.4	1701.1	5.5	2755.9
1977	18.3	1679.2	29.3	2722.6
1978	19.4	1659.0	31.1	2691.9
1979	6.5	1626.4	10.5	2641.2
1980	3.2	1591.1	5.1	2586.3
1981	8.1	1561.7	13.0	2540.6
1982	0.9	1525.7	1.5	2484.3
1983	0.9	1490.6	1.5	2429.4

Lake Michigan

1954	70.4	70.4	112.7	112.7
1955	91.1	159.9	145.8	255.9
1956	112.9	269.0	180.7	430.8
1957	113.3	376.0	181.3	602.2
1958	157.2	524.3	251.5	840.0
1959	221.3	733.1	354.0	1175.0
1960	40.0	755.8	64.1	1212.3
1961	62.0	800.0	99.1	1283.9
1962	314.6	1095.7	503.4	1758.2
1963	607.1	1676.9	971.4	2689.7
1964	404.5	2041.8	647.2	3275.8
1965	141.1	2134.7	225.7	3427.0
1966	61.8	2146.0	98.9	3448.1
1967	42.0	2137.3	67.1	3436.9
1968	33.6	2120.5	53.8	3412.7
1969	36.5	2106.9	58.4	3393.6
1970	37.7	2094.8	60.3	3376.8
1971	36.1	2081.5	57.8	3357.9
1972	19.2	2051.5	30.7	3312.3
1973	10.6	2013.6	17.0	3254.0
1974	23.8	1989.8	38.0	3218.1
1975	17.4	1960.2	27.8	3172.8
1976	3.9	1917.8	6.2	3106.9
1977	20.7	1893.2	33.1	3069.5
1978	21.9	1870.4	35.1	3034.8
1979	7.4	1833.5	11.8	2977.7
1980	3.6	1793.8	5.7	2915.8
1981	9.2	1760.6	14.7	2864.2
1982	1.0	1720.1	1.6	2800.8
1983	1.0	1680.5	1.6	2738.8

Lake Huron

1954	68.7	68.7	109.9	109.9
1955	88.8	155.8	142.1	249.5
1956	110.1	262.3	176.2	419.9
1957	110.4	366.5	176.7	587.1
1958	153.2	511.1	245.2	818.9
1959	215.7	714.7	345.1	1145.5
1960	39.0	736.9	62.5	1181.9
1961	60.4	779.9	96.6	1251.7
1962	306.7	1068.2	490.8	1714.0
1963	591.9	1634.8	947.0	2622.1
1964	394.3	1990.6	630.9	3193.5
1965	137.5	2081.1	220.0	3341.0
1966	60.3	2092.2	96.4	3361.5
1967	40.9	2083.7	65.5	3350.6
1968	32.8	2067.2	52.5	3327.0
1969	35.6	2054.0	57.0	3308.4

1970	36.8	2042.2	58.8	3292.0
1971	35.2	2029.2	56.3	3273.6
1972	18.7	2000.0	29.9	3229.1
1973	10.4	1963.1	16.6	3172.3
1974	23.2	1939.9	37.1	3137.3
1975	16.9	1911.0	27.1	3093.2
1976	3.8	1869.6	6.0	3028.9
1977	20.2	1845.6	32.2	2992.4
1978	21.4	1823.4	34.2	2958.6
1979	7.2	1787.5	11.5	2902.9
1980	3.5	1748.8	5.6	2842.6
1981	8.9	1716.4	14.3	2792.3
1982	1.0	1676.9	1.6	2730.5
1983	1.0	1638.3	1.6	2670.1

Lake Erie

1954	73.5	73.5	117.6	117.6
1955	95.1	166.9	152.2	267.1
1956	117.9	280.8	188.6	449.7
1957	118.3	392.5	189.2	628.7
1958	164.1	547.3	262.5	877.0
1959	231.0	765.3	369.6	1226.6
1960	41.8	789.1	66.9	1265.6
1961	64.7	835.1	103.5	1340.3
1962	328.5	1143.8	525.5	1835.4
1963	633.8	1750.6	1014.1	2807.9
1964	422.3	2131.5	675.6	3419.7
1965	147.3	2228.5	235.6	3577.6
1966	64.5	2240.3	103.2	3599.6
1967	43.8	2231.2	70.1	3587.9
1968	35.1	2213.6	56.2	3562.6
1969	38.1	2199.5	61.0	3542.7
1970	39.4	2186.9	63.0	3525.2
1971	37.7	2172.9	60.3	3505.4
1972	20.0	2141.6	32.0	3457.8
1973	11.1	2102.1	17.7	3397.0
1974	24.8	2077.3	39.7	3359.6
1975	18.1	2046.3	29.0	3312.3
1976	4.0	2002.0	6.5	3243.5
1977	21.6	1976.3	34.5	3204.3
1978	22.9	1952.5	36.6	3168.2
1979	7.7	1914.1	12.3	3108.5
1980	3.7	1872.6	6.0	3043.9
1981	9.6	1838.0	15.3	2990.1
1982	1.1	1795.7	1.7	2923.9
1983	1.1	1754.3	1.7	2859.2

Lake Ontario

1954	71.3	71.3	114.1	114.1
1955	92.2	161.9	147.6	259.1
1956	114.4	272.4	183.0	436.2
1957	114.7	380.7	183.5	609.8
1958	159.2	530.8	254.6	850.6
1959	224.0	742.3	358.5	1189.7
1960	40.5	765.3	64.9	1227.6
1961	62.7	810.0	100.4	1300.0
1962	318.6	1109.4	509.7	1780.2
1963	614.8	1698.0	983.6	2723.4
1964	409.6	2067.5	655.3	3316.9
1965	142.8	2161.5	228.5	3470.1
1966	62.6	2173.0	100.1	3491.4
1967	42.5	2164.2	68.0	3480.1
1968	34.1	2147.1	54.5	3455.5
1969	37.0	2133.4	59.2	3436.2
1970	38.2	2121.1	61.1	3419.2
1971	36.6	2107.6	58.5	3400.0
1972	19.4	2077.2	31.0	3353.9
1973	10.7	2038.9	17.2	3294.9
1974	24.1	2014.8	38.5	3258.6
1975	17.6	1984.8	28.1	3212.7
1976	3.9	1941.9	6.3	3146.0
1977	20.9	1916.9	33.5	3108.0
1978	22.2	1893.8	35.5	3072.9
1979	7.5	1856.6	12.0	3015.1
1980	3.6	1816.3	5.8	2952.4
1981	9.3	1782.7	14.9	2900.2
1982	1.1	1741.7	1.7	2836.0
1983	1.1	1701.6	1.7	2773.2

CAPTIONS FOR FIGURES

Fig. 1. Nuclear facilities in the Great Lakes basin.

Fig. 2. The growth of nuclear power production in the Great Lakes basin.

Fig. 3. Latitudinal dependence of the deposition of fallout ^{90}Sr.

Fig. 4. Estimated inputs and current (1989) inventories of ^{90}Sr in the Great Lakes basin.

Fig. 5. Estimated inputs and current (1989) inventories of ^{137}Cs in the Great Lakes basin.

Fig. 6. Estimated inputs of ^{3}H to the Great Lakes.

Fig. 7. Distribution of ^{137}Cs in Lake Ontario waters, surface sediments and biota.

Fig. 8. Specific activities of ^{137}Cs, ^{226}Ra, (^{238}Pu + $^{239,240}Pu$) and ^{241}Am in various compartments in Lake Ontario.

Fig. 9. Distribution of ^{137}Cs and $^{239,240}Pu$ in various particle size classes of Lake Michigan surface sediments.

Fig. 10. Seasonal cycling of $^{239,240}Pu$ in Lake Michigan.

Fig. 11. Levels of representative radionuclides in Great Lakes waters.

Fig. 12. Distribution of ^{137}Cs in Great Lakes sediments.

Fig. 13. Distribution of $^{239,240}Pu$ in the sediments of Lakes Michigan and Ontario.

Fig. 14. Concentration factors for ^{137}Cs in Great Lakes biota.

Fig. 15. Concentration factors for $^{239,240}Pu$ in biota from Lakes Michigan and Ontario.

Fig. 16. Distribution of ^{137}Cs and $^{239,240}Pu$ in different organs of Lake Ontario fish.

Fig. 17. Comparison of model-predicted and measured concentrations of ^{90}Sr and $^{239,240}Pu$ in Great Lakes waters.

Fig. 18. Average annual concentrations of ^{226}Ra in the Serpent River. Data from IJC (1989).

Fig. 19. Levels of ^{90}Sr in Cattaraugus Creek (NYSDEC, 1967-81; NYSDH, 1982-86).

Fig. 20. $^{239,240}Pu$ profile in a ^{210}Pb-dated sediment core from Lake Ontario near the mouth of Niagara River. The arbitrary time scale is used only to approximate the deposition of direct fallout (over Lake Ontario) and West Valley and Lake Erie-supplied $^{239,240}Pu$.

Fig. 21. Model-predicted concentrations of ^{226}Ra at the Port Granby radioactive waste management site following relocation of the waste.

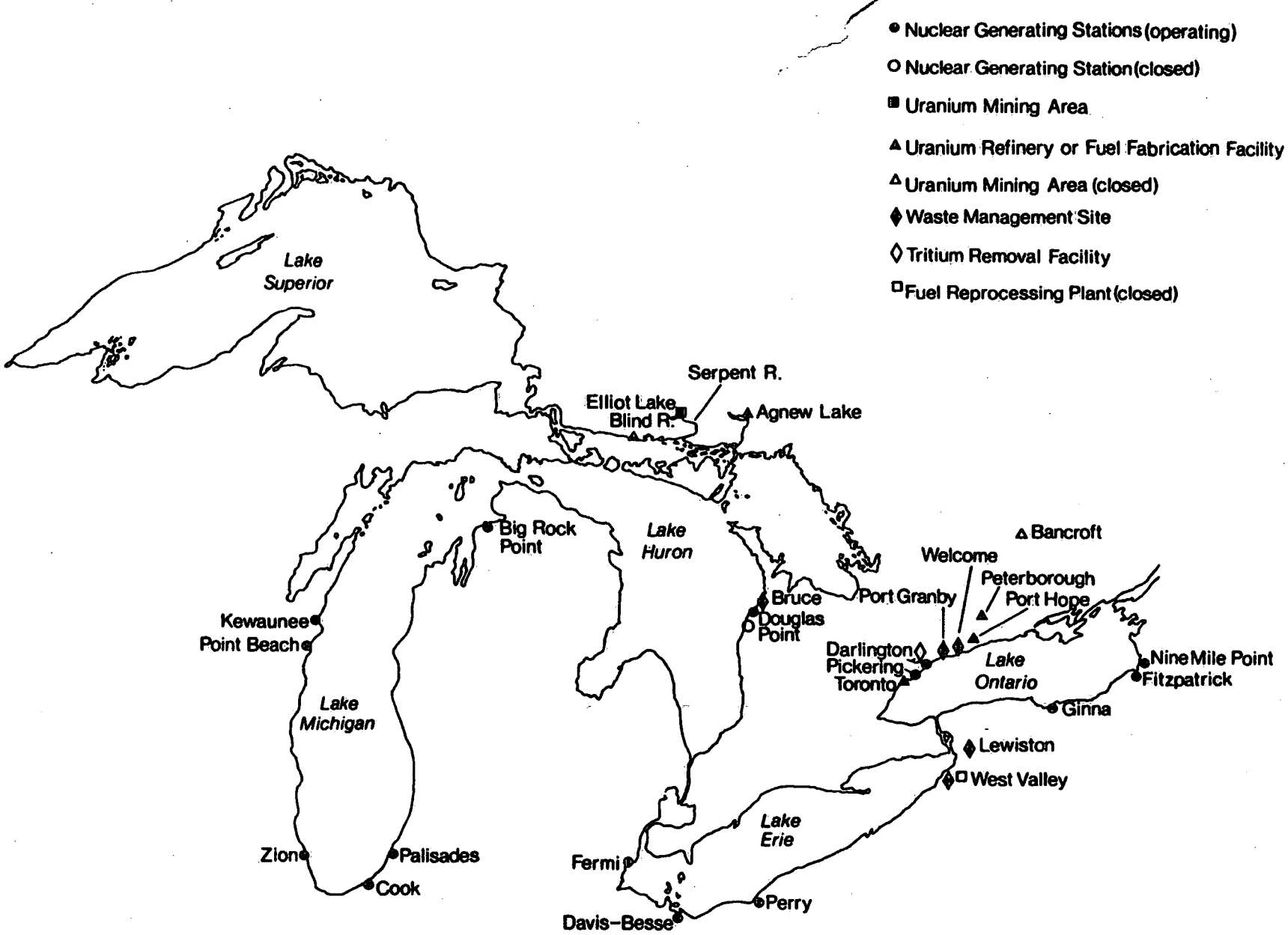

Nuclear Generating Stations (operating)
Nuclear Generating Station (closed)
Uranium Mining Area
Uranium Refinery or Fuel Fabrication Facility
Uranium Mining Area (closed)
Waste Management Site
Tritium Removal Facility
Fuel Reprocessing Plant (closed)
Lake Superior
Serpent R.
Elliot Lake
Blind R.
Agnew Lake
Big Rock Point
Lake Huron
Bancroft
Welcome
Peterborough
Port Hope
Bruce
Port Granby
Douglas Point
Kewaunee
Point Beach
Darlington
Pickering
Toronto
Lake Ontario
Nine Mile Point
Fitzpatrick
Ginna
Lake Michigan
Lewiston
West Valley
Lake Erie
Zion
Palisades
Cook
Fermi
Perry
Davis-Besse

FIGURE 1

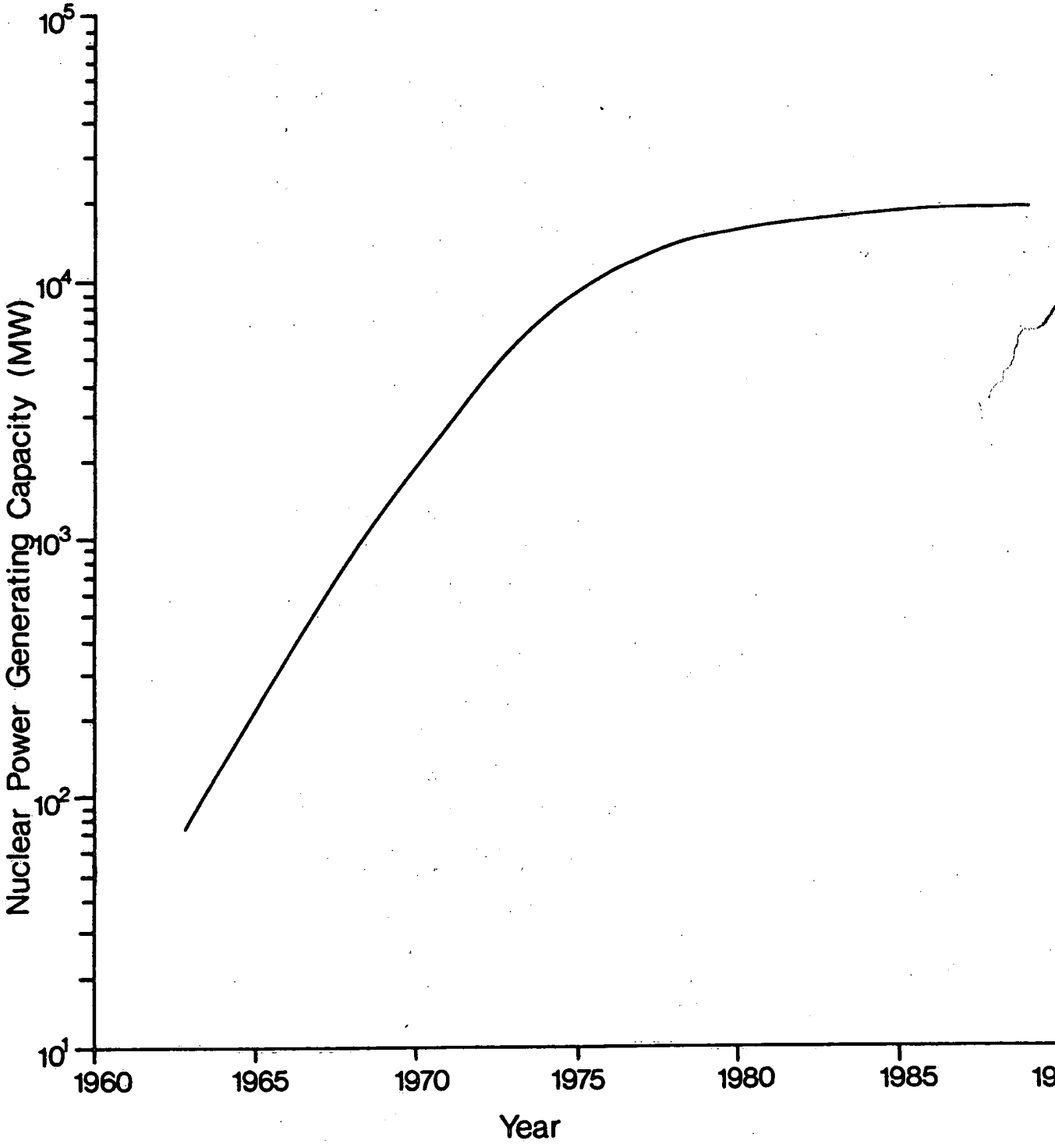

10^5
10^4
10^3
10^2
10^1
Nuclear Power Generating Capacity (MW)
1960
1965
1970
1975
1980
1985
Year

FIGURE 2

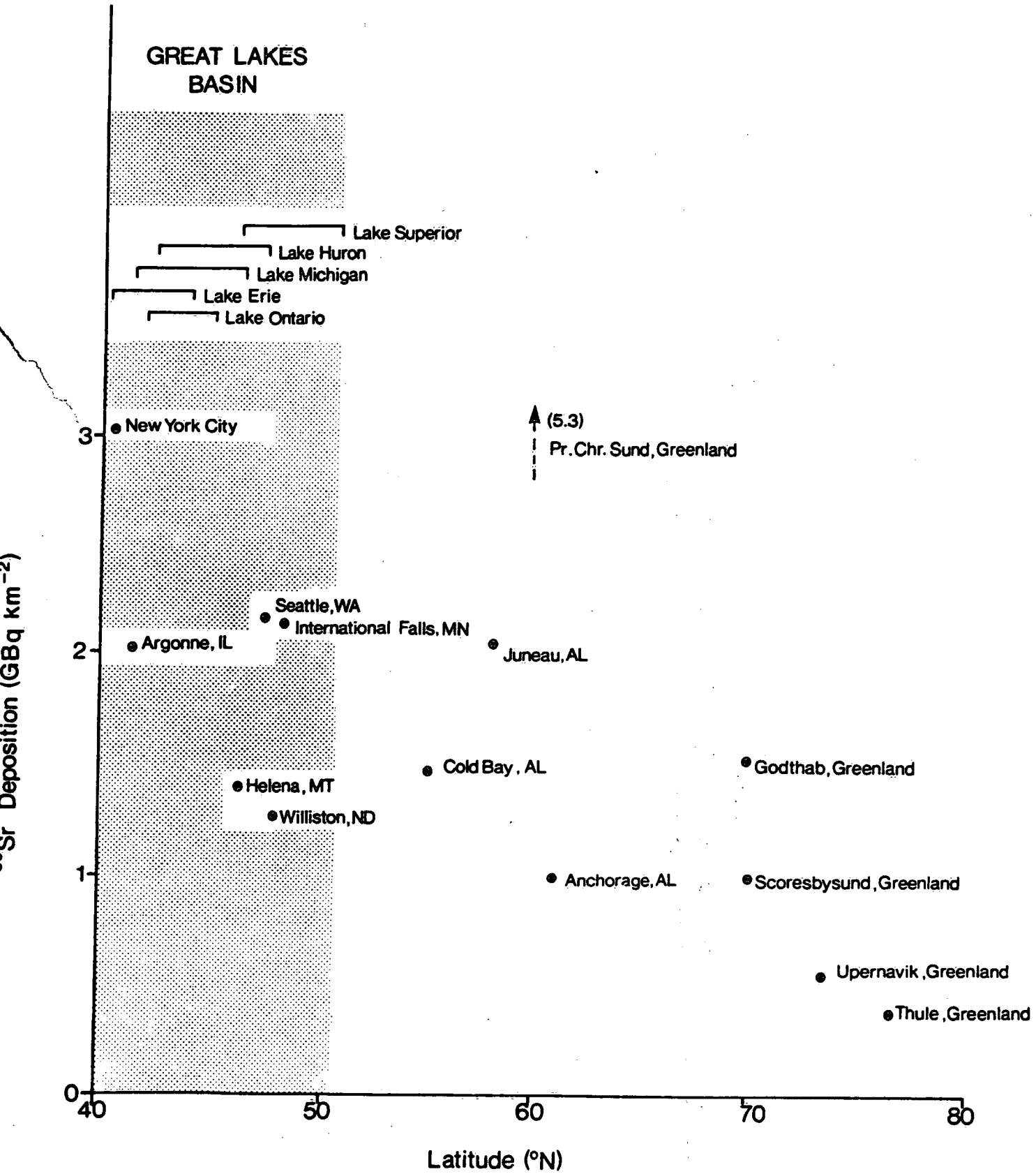

GREAT LAKES BASIN
Lake Superior
Lake Huron
Lake Michigan
Lake Erie
Lake Ontario
New York City
(5.3)
Pr. Chr. Sund, Greenland
Seattle, WA
International Falls, MN
Argonne, IL
Juneau, AL
Cold Bay, AL
Godthab, Greenland
Helena, MT
Williston, ND
Anchorage, AL
Scoresbysund, Greenland
Upernavik, Greenland
Thule, Greenland
^{90}Sr Deposition (GBq km^{-2})
0
1
2
3
40
50
60
70
80
Latitude (°N)

FIGURE 3

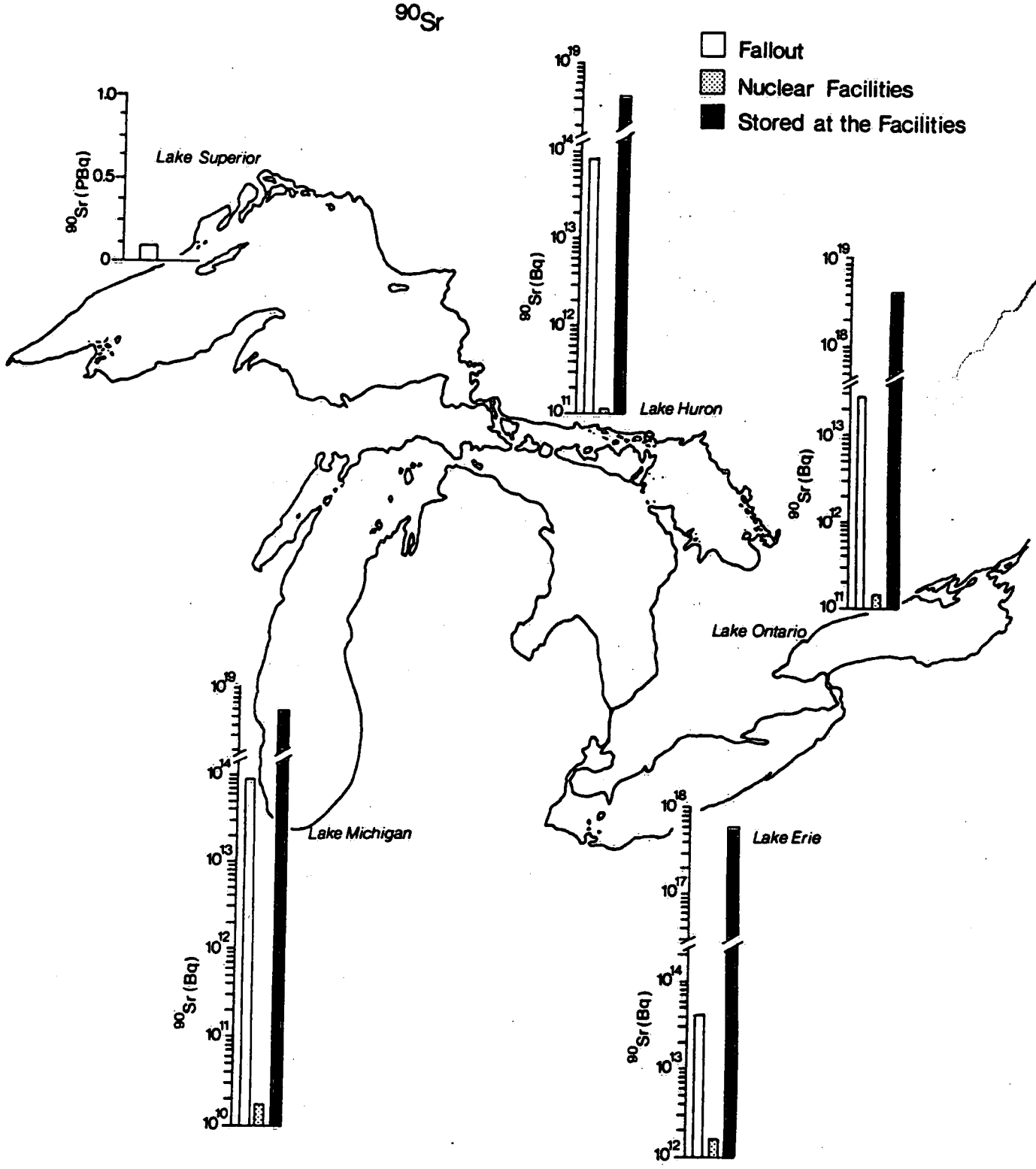

^{90}Sr
Fallout
Nuclear Facilities
Stored at the Facilities
Lake Superior
^{90}Sr (PBq)
1.0
0.5
0
Lake Huron
Lake Michigan
Lake Ontario
Lake Erie
^{90}Sr (Bq)

FIGURE 4

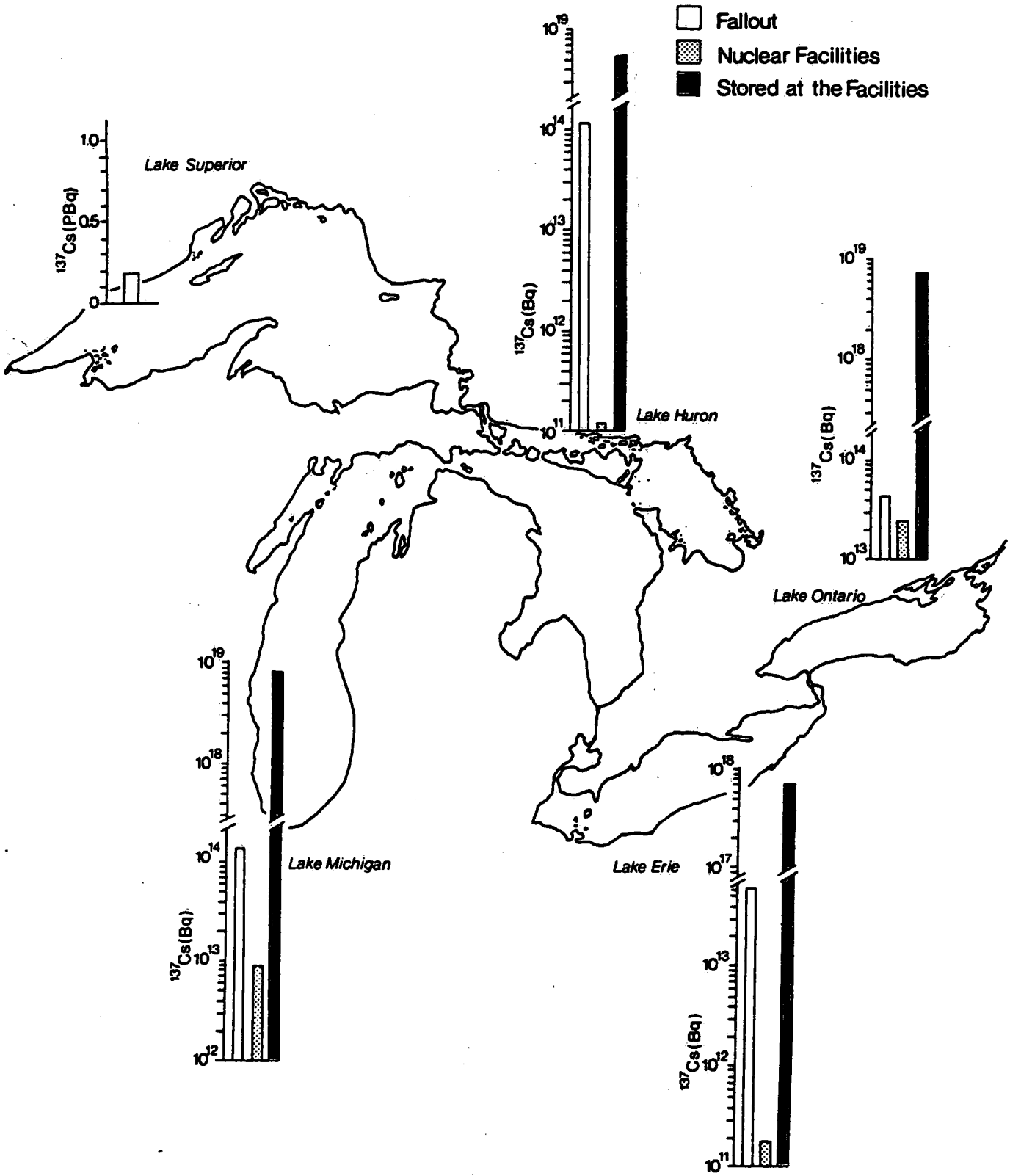

137Cs
Fallout
Nuclear Facilities
Stored at the Facilities
Lake Superior
137Cs (PBq)
1.0
0.5
0
Lake Huron
137Cs (Bq)
Lake Ontario
Lake Michigan
Lake Erie

FIGURE 5

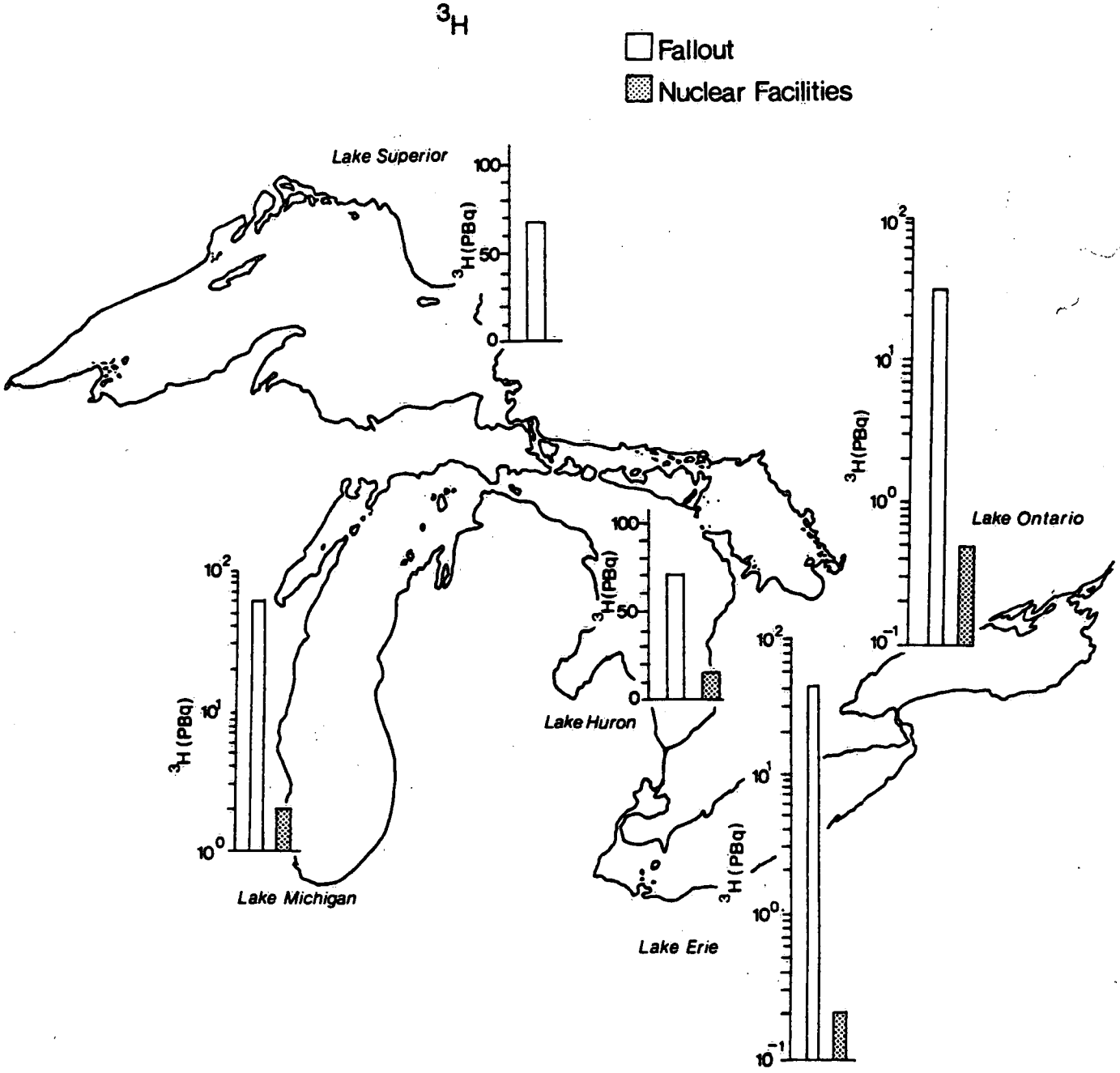

^3H
Fallout
Nuclear Facilities
Lake Superior
^3H (PBq)
100
50
0
Lake Michigan
10^2
10^1
10^0
Lake Huron
Lake Erie
Lake Ontario
10^{-1}

FIGURE 6

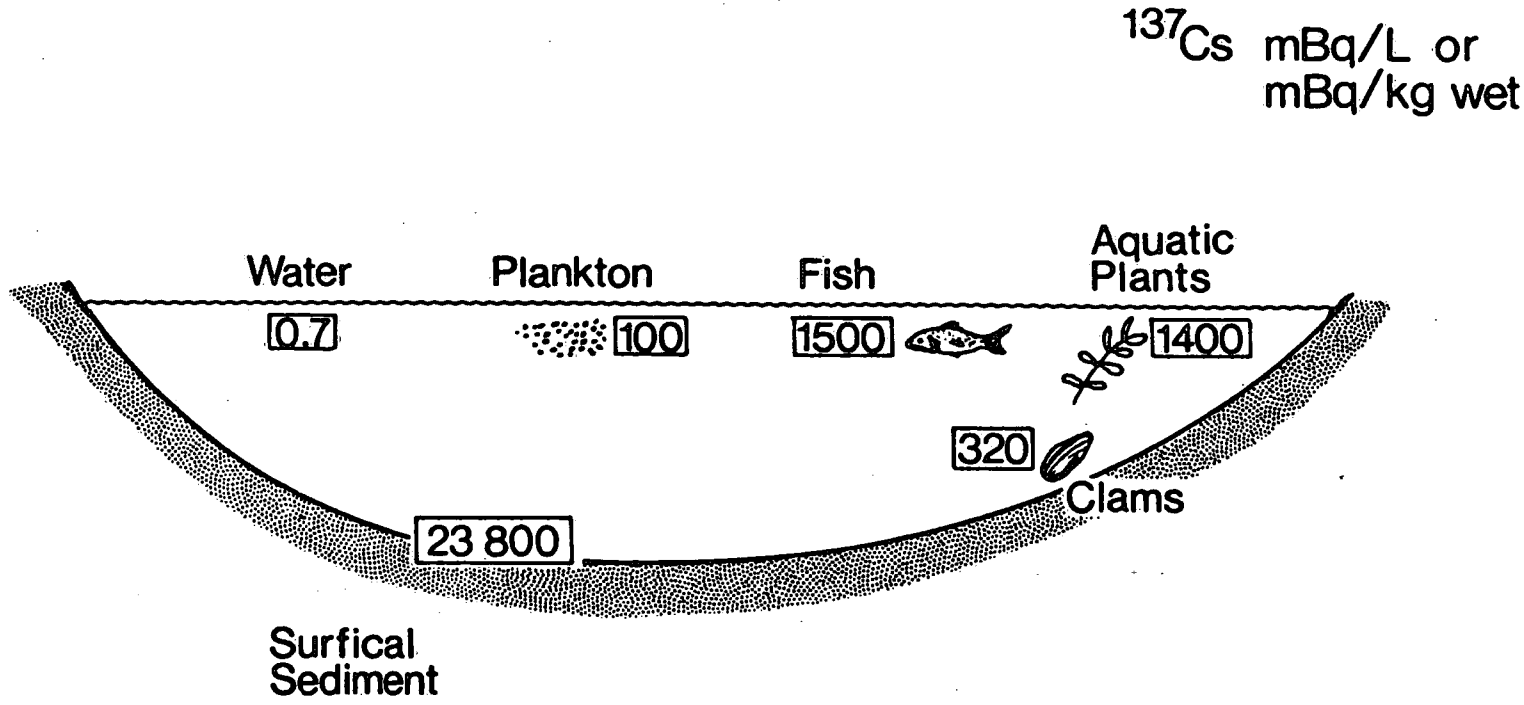
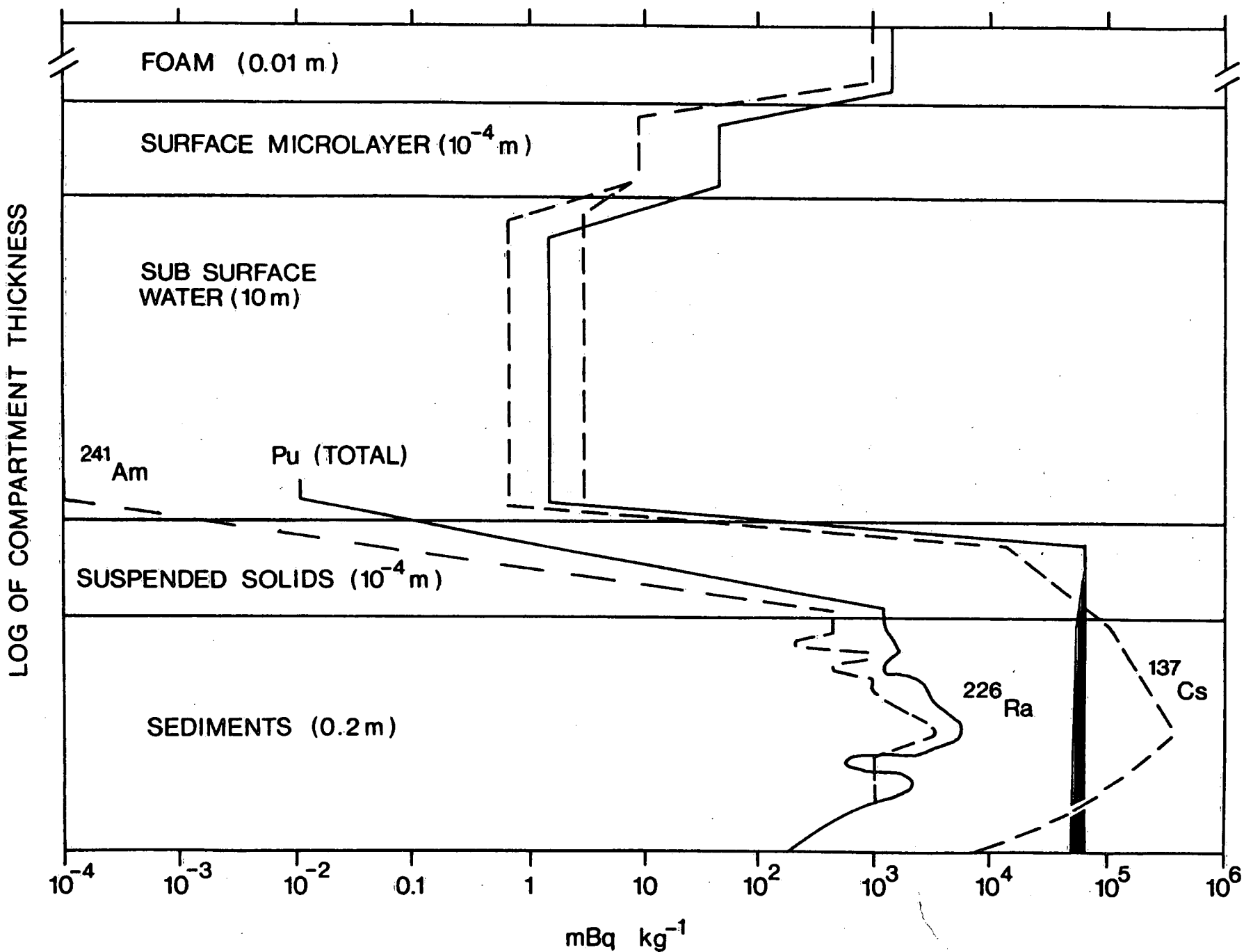

^{137}Cs mBq/L or
mBq/kg wet
Water
Plankton
Fish
Aquatic
Plants
0.7
100
1500
1400
320
Clams
23 800
Surfical
Sediment

FIGURE 7


FOAM (0.01 m)
SURFACE MICROLAYER (10^{-4} m)
SUB SURFACE WATER (10 m)
SUSPENDED SOLIDS (10^{-4} m)
SEDIMENTS (0.2 m)
LOG OF COMPARTMENT THICKNESS
^{241}Am
Pu (TOTAL)
^{226}Ra
^{137}Cs
10^{-4}
10^{-3}
10^{-2}
0.1
1
10
10^{2}
10^{3}
10^{4}
10^{5}
10^{6}
mBq kg^{-1}

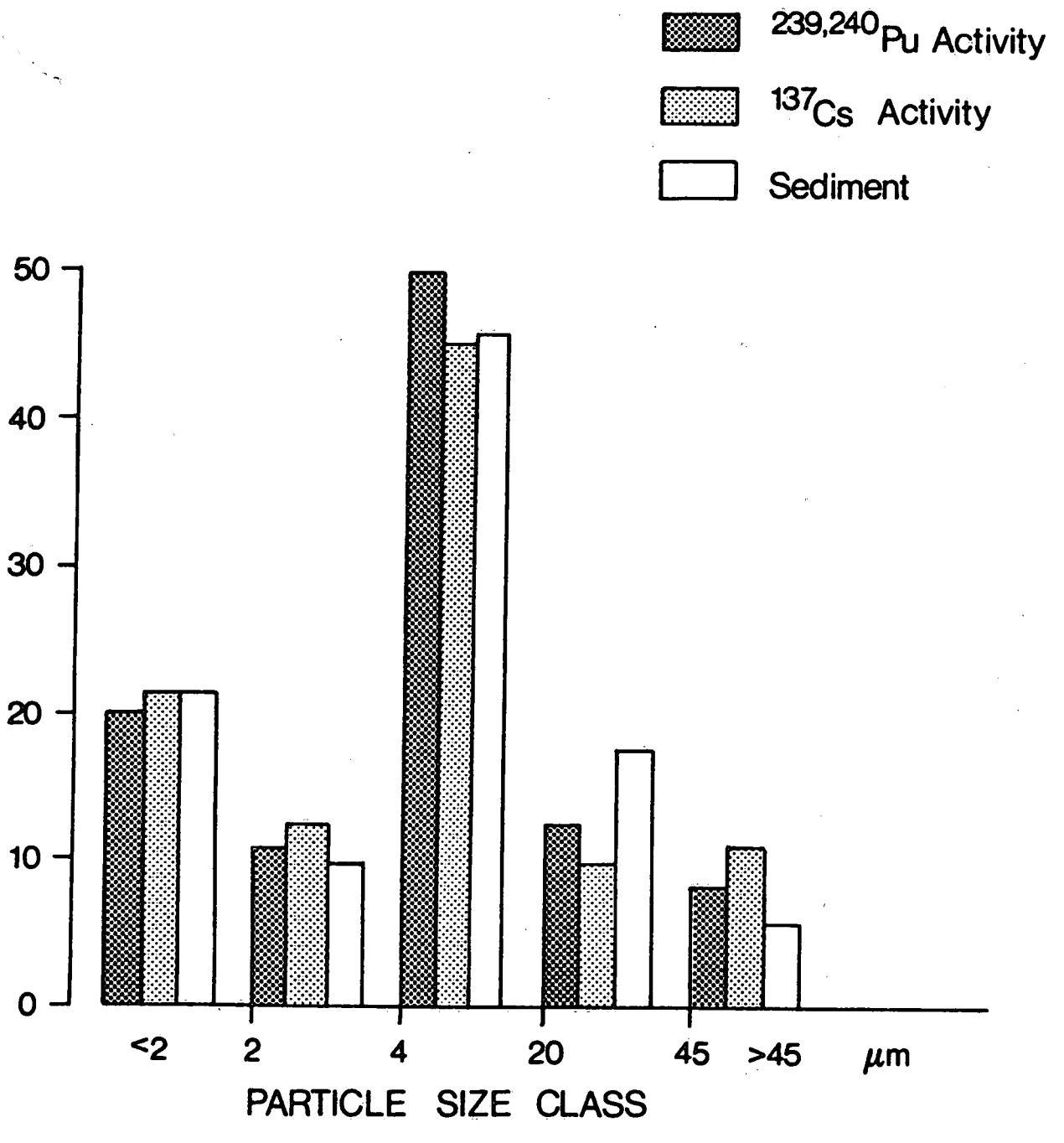

239,240Pu Activity
137Cs Activity
Sediment
50
40
30
20
10
0
<2
2
4
20
45
>45
μm
PARTICLE SIZE CLASS

FIGURE 9

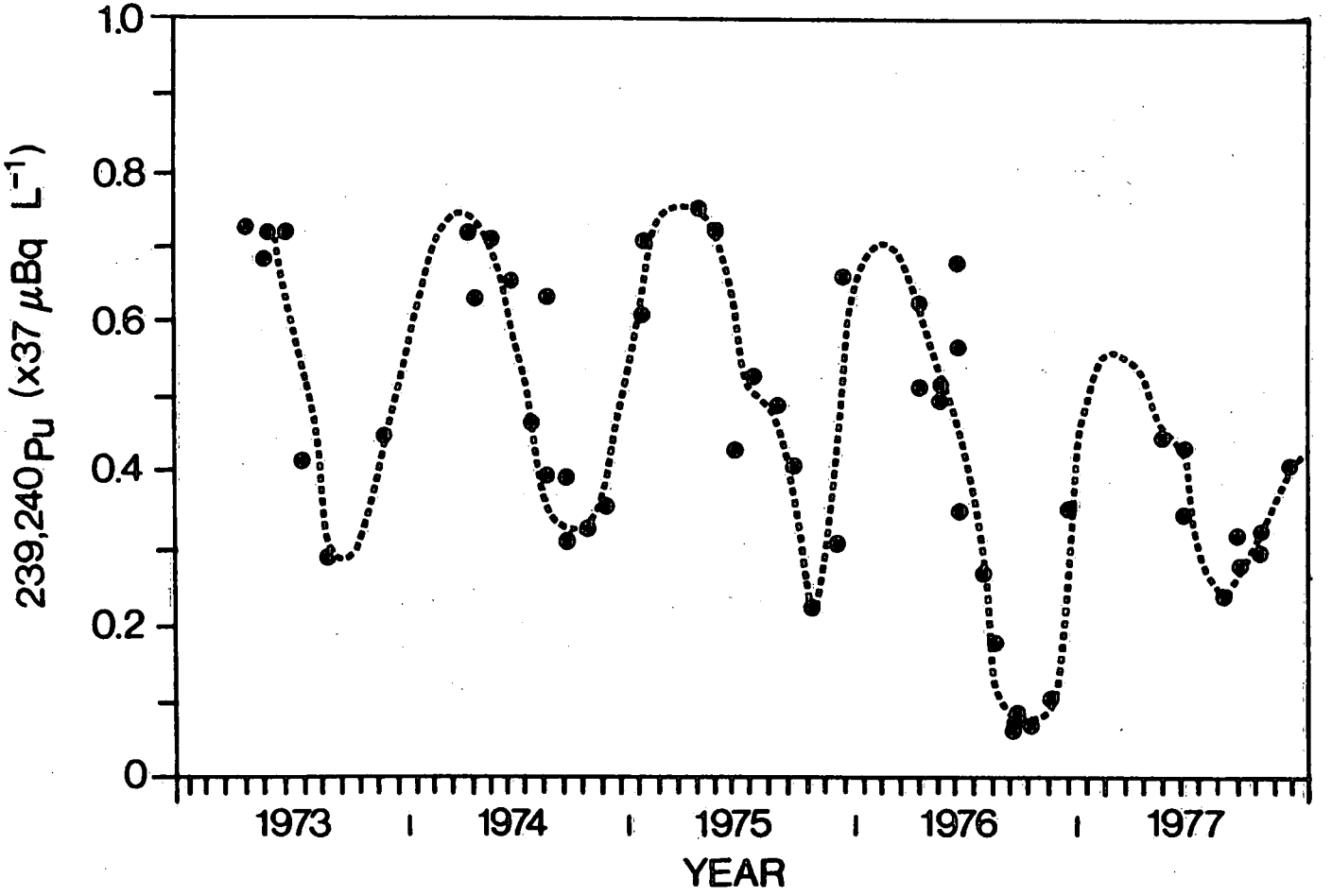

1.0
0.8
0.6
0.4
0.2
0
239,240Pu (x37 μBq L-1)
1973
1974
1975
1976
1977
YEAR

FIGURE 10

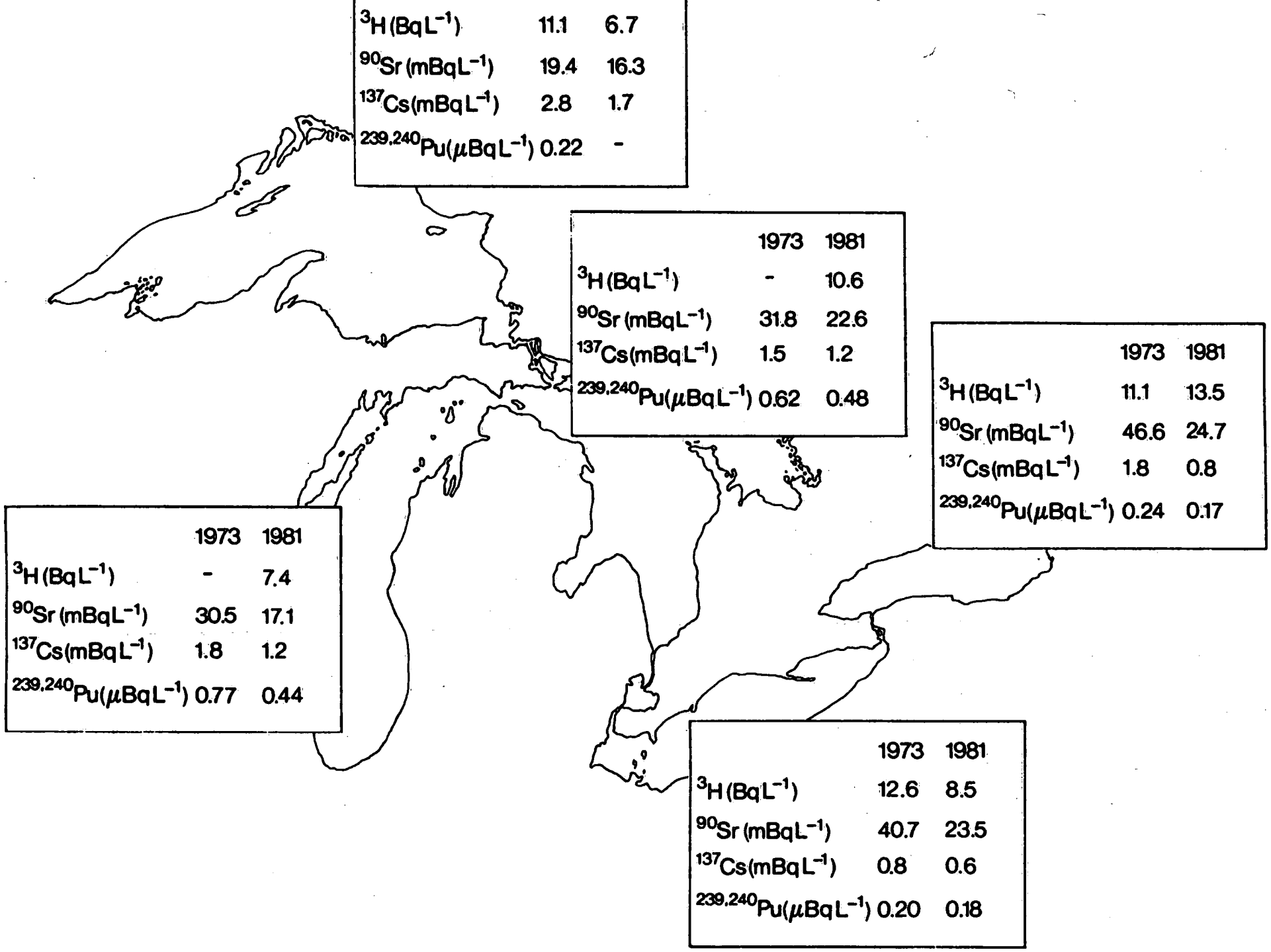

^{3}H (BqL^{-1}) 11.1 6.7
^{90}Sr (mBqL^{-1}) 19.4 16.3
^{137}Cs (mBqL^{-1}) 2.8 1.7
$^{239,240}Pu$ (μBqL^{-1}) 0.22 -
1973 1981
^{3}H (BqL^{-1}) - 10.6
^{90}Sr (mBqL^{-1}) 31.8 22.6
^{137}Cs (mBqL^{-1}) 1.5 1.2
$^{239,240}Pu$ (μBqL^{-1}) 0.62 0.48
1973 1981
^{3}H (BqL^{-1}) 11.1 13.5
^{90}Sr (mBqL^{-1}) 46.6 24.7
^{137}Cs (mBqL^{-1}) 1.8 0.8
$^{239,240}Pu$ (μBqL^{-1}) 0.24 0.17
1973 1981
^{3}H (BqL^{-1}) - 7.4
^{90}Sr (mBqL^{-1}) 30.5 17.1
^{137}Cs (mBqL^{-1}) 1.8 1.2
$^{239,240}Pu$ (μBqL^{-1}) 0.77 0.44
1973 1981
^{3}H (BqL^{-1}) 12.6 8.5
^{90}Sr (mBqL^{-1}) 40.7 23.5
^{137}Cs (mBqL^{-1}) 0.8 0.6
$^{239,240}Pu$ (μBqL^{-1}) 0.20 0.18

FIGURE 11

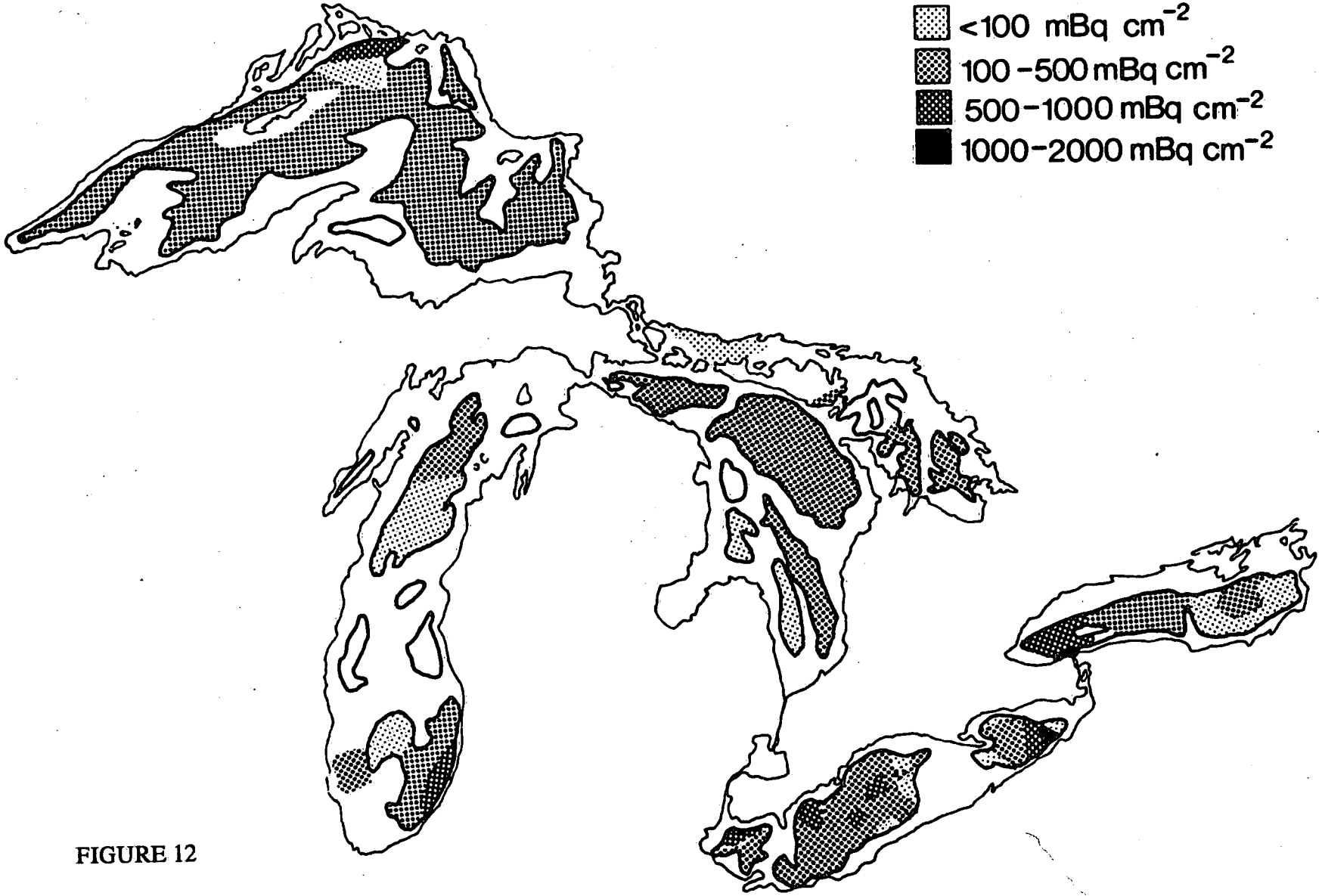


FIGURE 12

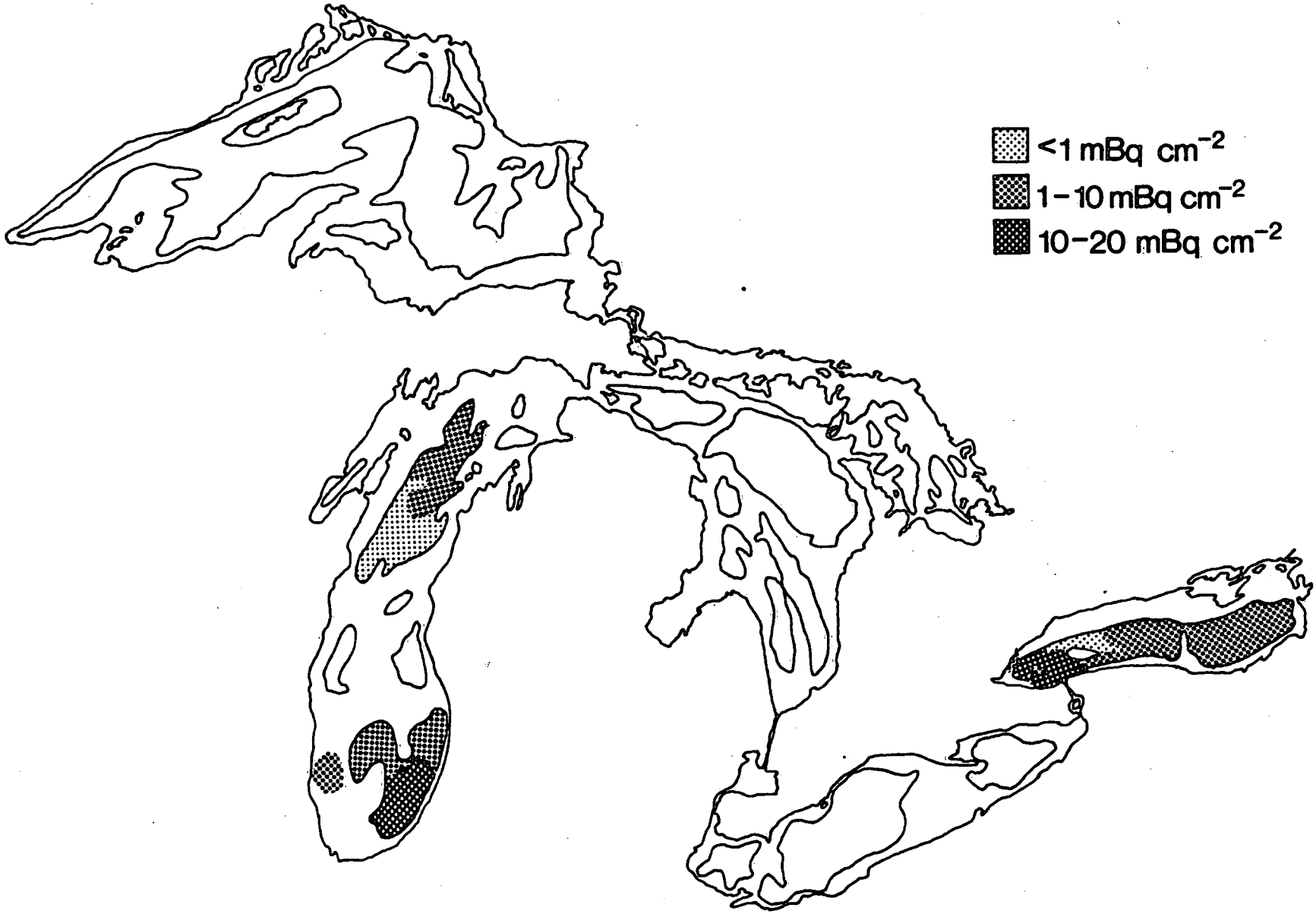


FIGURE 13

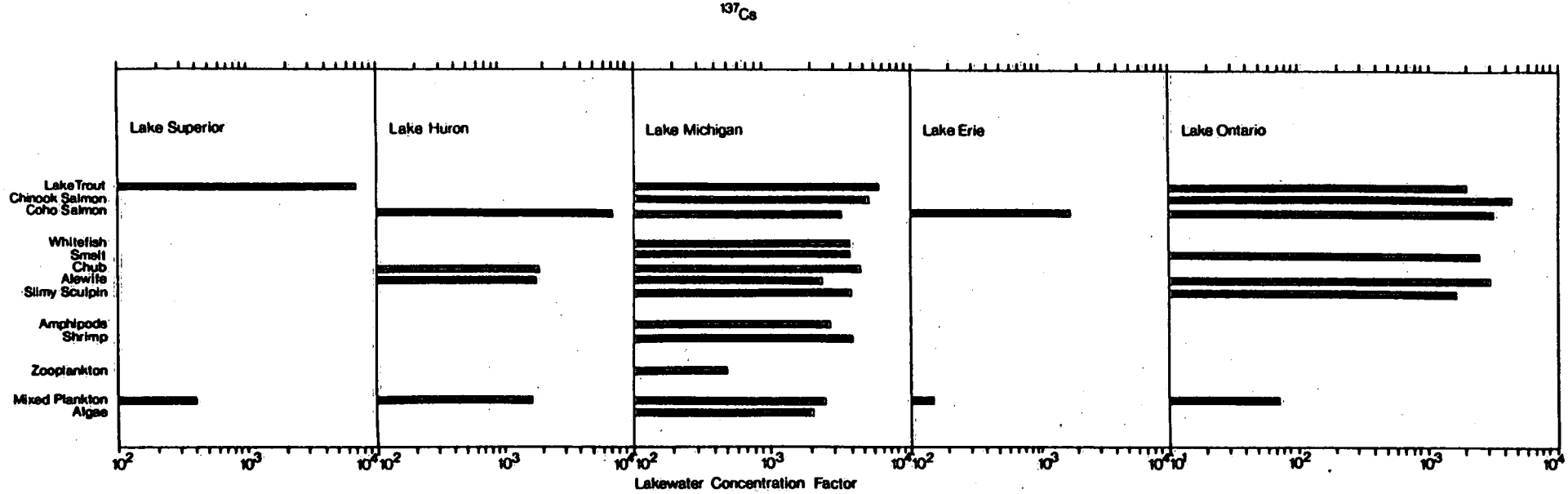

^{137}Cs
Lake Superior
Lake Huron
Lake Michigan
Lake Erie
Lake Ontario
Lake Trout
Chinook Salmon
Coho Salmon
Whitefish
Smelt
Chub
Alewife
Slimy Sculpin
Amphipods
Shrimp
Zooplankton
Mixed Plankton
Algae
10^2
10^3
10^4
Lakewater Concentration Factor

FIGURE 14

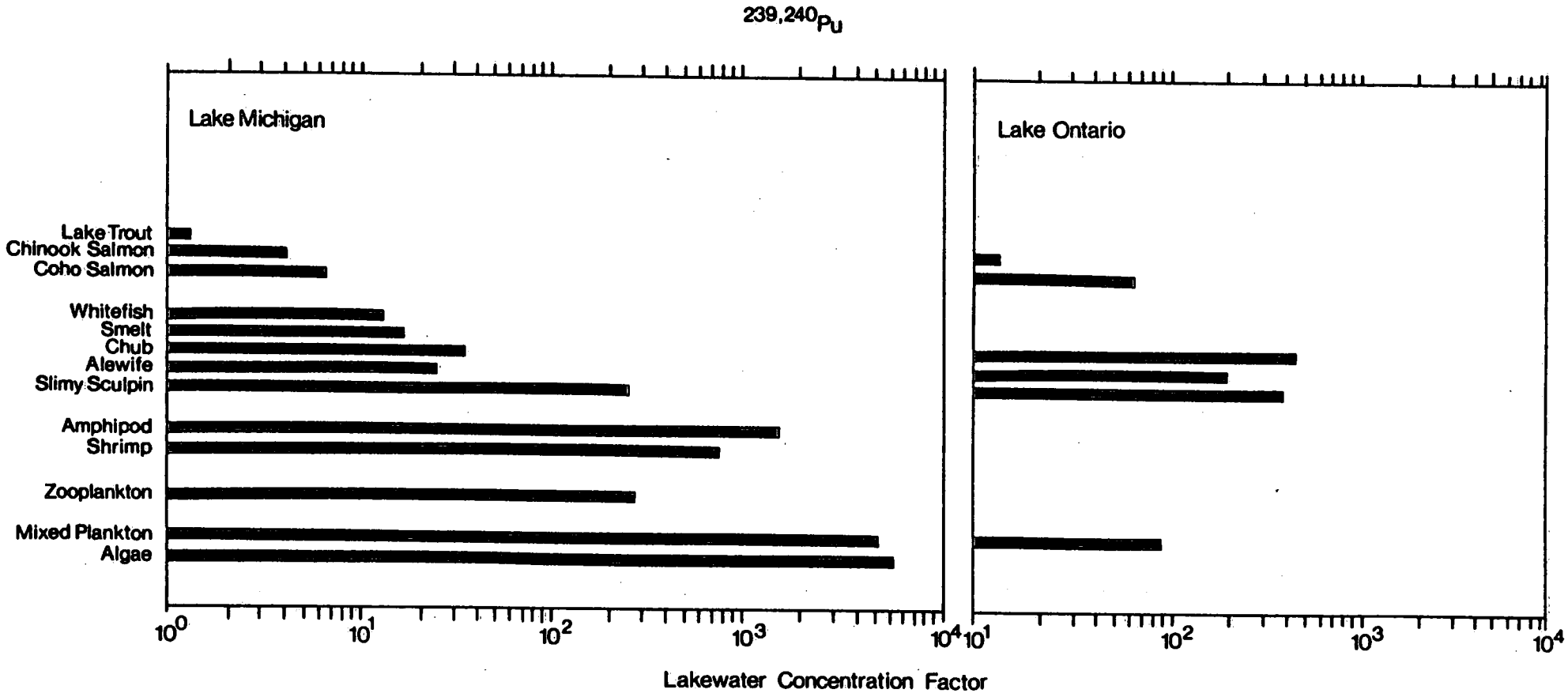

239,240Pu
Lake Michigan
Lake Ontario
Lake Trout
Chinook Salmon
Coho Salmon
Whitefish
Smelt
Chub
Alewife
Slimy Sculpin
Amphipod
Shrimp
Zooplankton
Mixed Plankton
Algae
10^0
10^1
10^2
10^3
10^4
10^1
10^2
10^3
10^4
Lakewater Concentration Factor

FIGURE 15

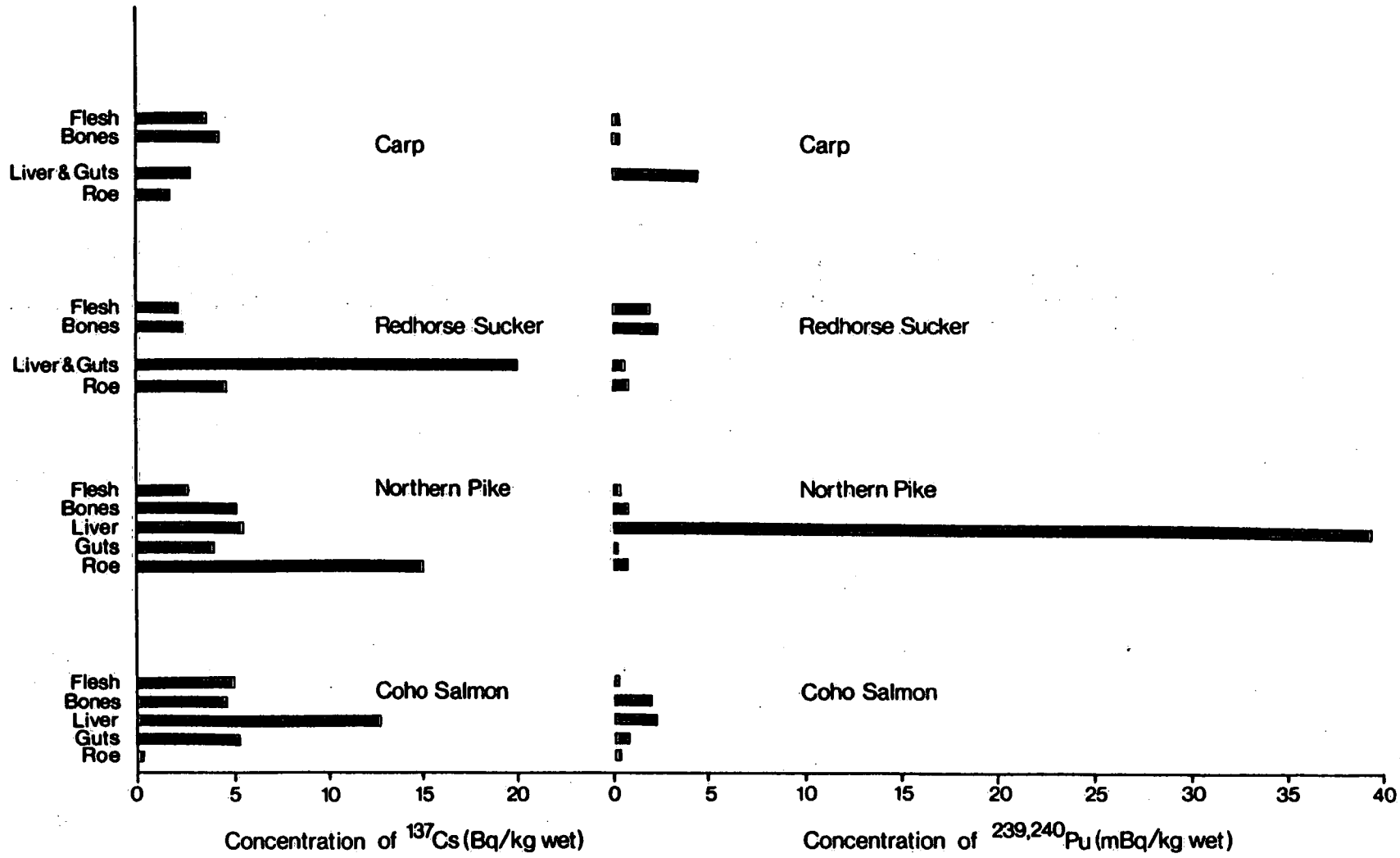

Flesh
Bones
Liver & Guts
Roe
Carp
Carp
Flesh
Bones
Liver & Guts
Roe
Redhorse Sucker
Redhorse Sucker
Flesh
Bones
Liver
Guts
Roe
Northern Pike
Northern Pike
Flesh
Bones
Liver
Guts
Roe
Coho Salmon
Coho Salmon
0
5
10
15
20
0
5
10
15
20
25
30
35
40
Concentration of 137Cs (Bq/kg wet)
Concentration of 239,240Pu (mBq/kg wet)

FIGURE 16

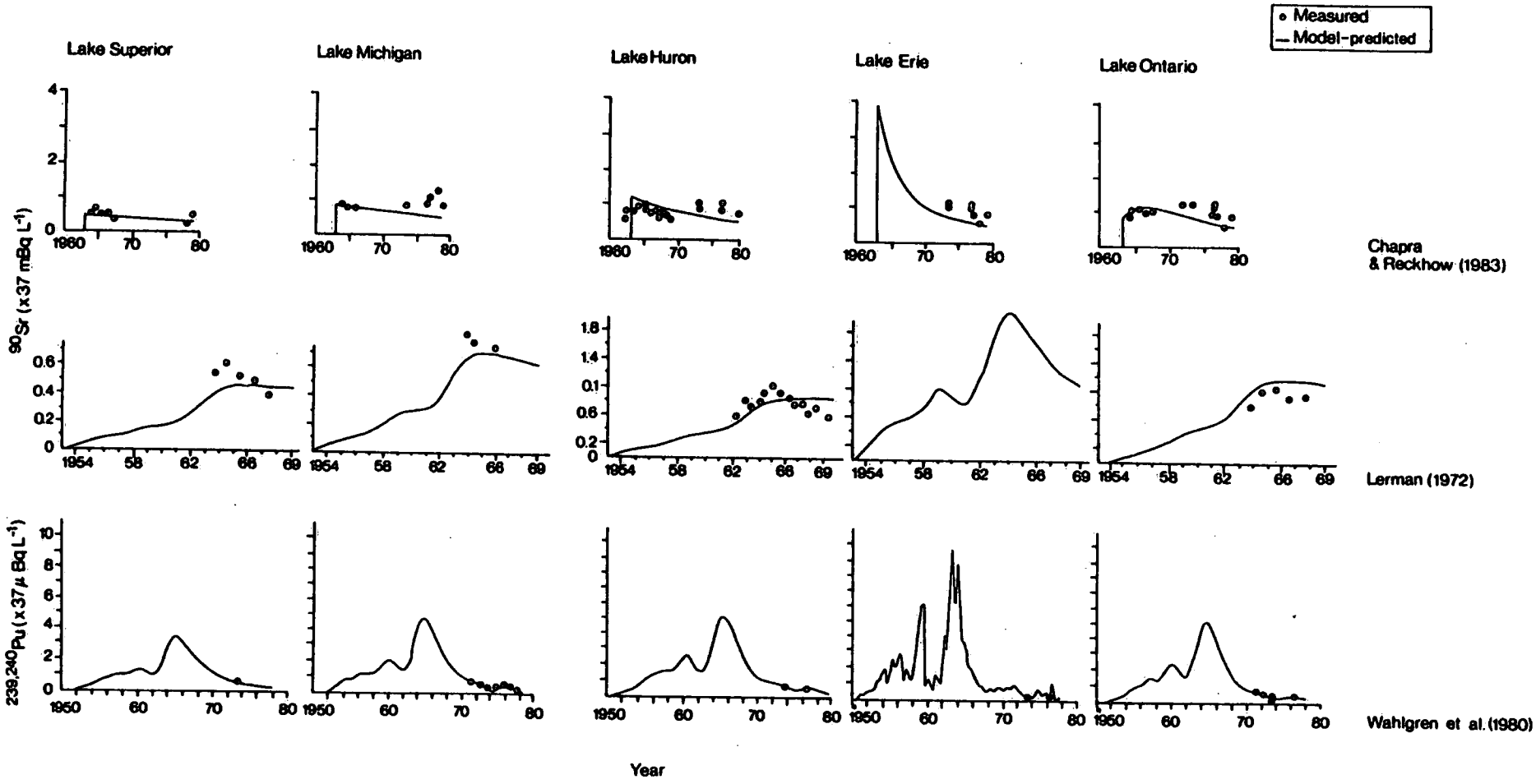

Measured
Model-predicted
Lake Superior
Lake Michigan
Lake Huron
Lake Erie
Lake Ontario
^{90}Sr (x37 mBq L^{-1})
$^{239,240}Pu$ (x37 μ Bq L^{-1})
Year
Chapra & Reckhow (1983)
Lerman (1972)
Wahlgren et al. (1980)

FIGURE 17

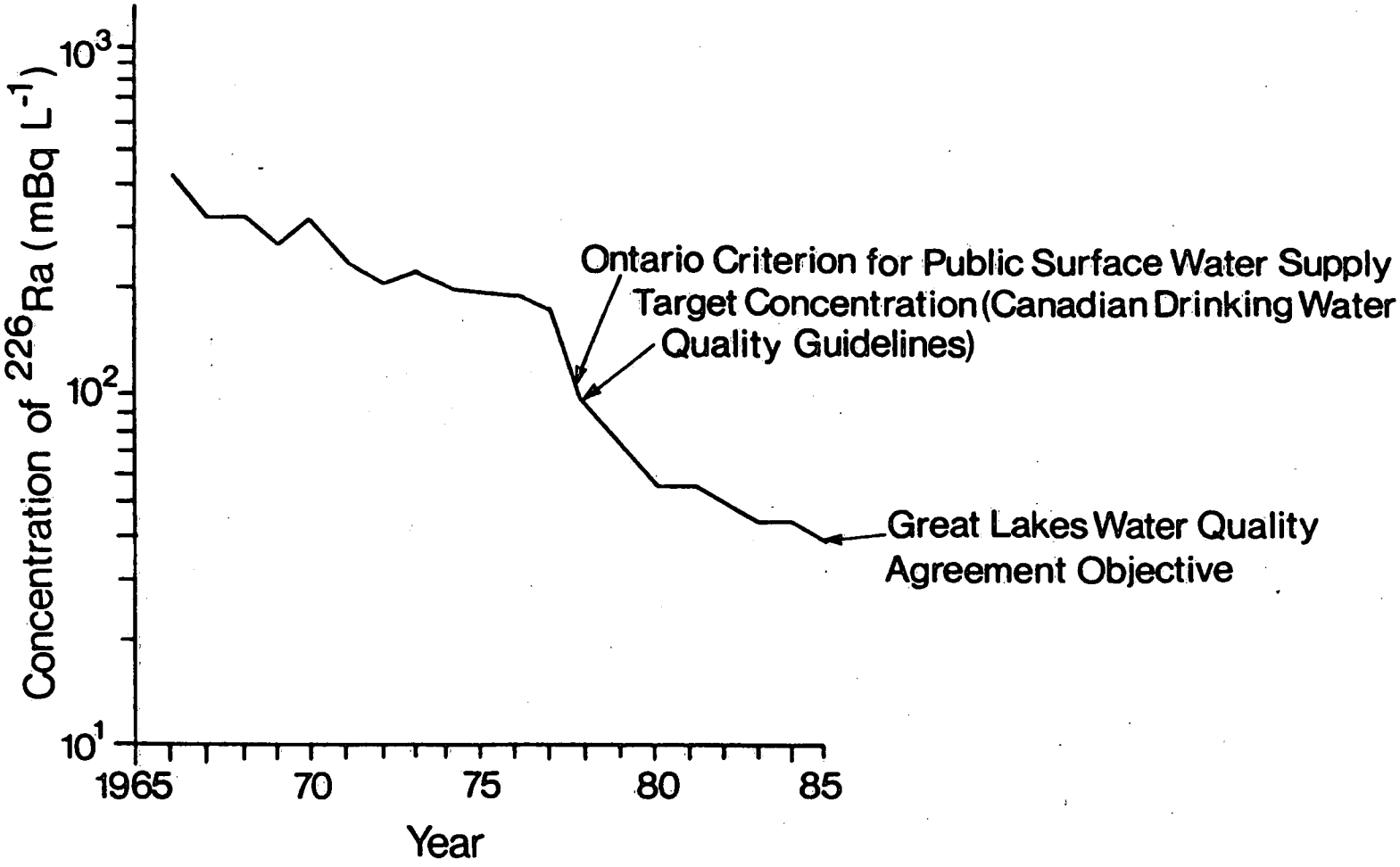

Concentration of ^{226}Ra (mBq L^{-1})
10^3
10^2
10^1
1965
70
75
80
85
Year
Ontario Criterion for Public Surface Water Supply
Target Concentration (Canadian Drinking Water
Quality Guidelines)
Great Lakes Water Quality
Agreement Objective

FIGURE 18

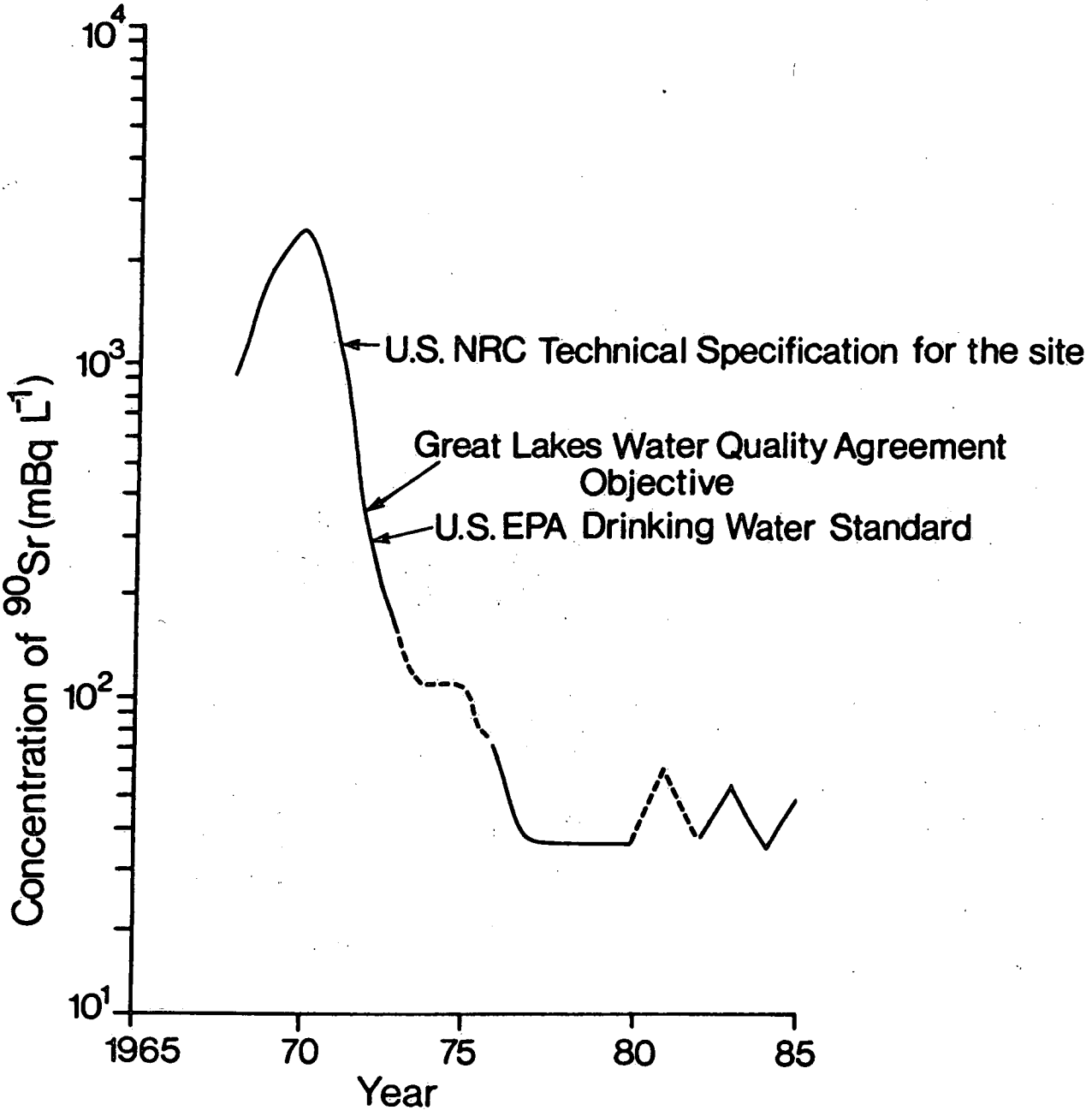

10^4
10^3
10^2
10^1
Concentration of ^{90}Sr (mBq L^{-1})
U.S. NRC Technical Specification for the site
Great Lakes Water Quality Agreement Objective
U.S. EPA Drinking Water Standard
1965
70
75
80
85
Year

FIGURE 19

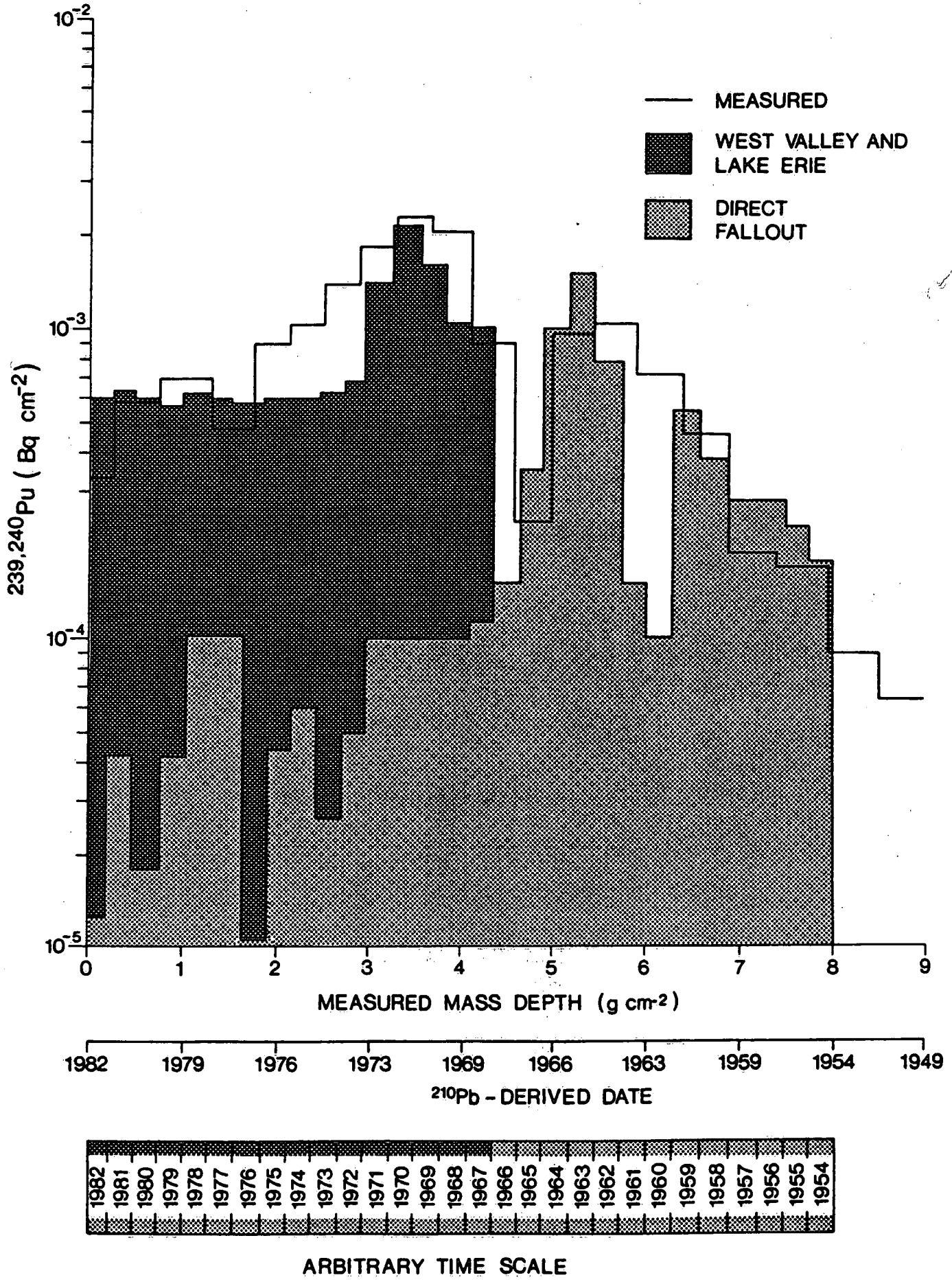

MEASURED
WEST VALLEY AND LAKE ERIE
DIRECT FALLOUT
239,240Pu (Bq cm-2)
10-2
10-3
10-4
10-5
0
1
2
3
4
5
6
7
8
9
MEASURED MASS DEPTH (g cm-2)
1982
1979
1976
1973
1969
1966
1963
1959
1954
1949
210Pb - DERIVED DATE
1982 1981 1980 1979 1978 1977 1976 1975 1974 1973 1972 1971 1970 1969 1968 1967 1966 1965 1964 1963 1962 1961 1960 1959 1958 1957 1956 1955 1954
ARBITRARY TIME SCALE

FIGURE 20

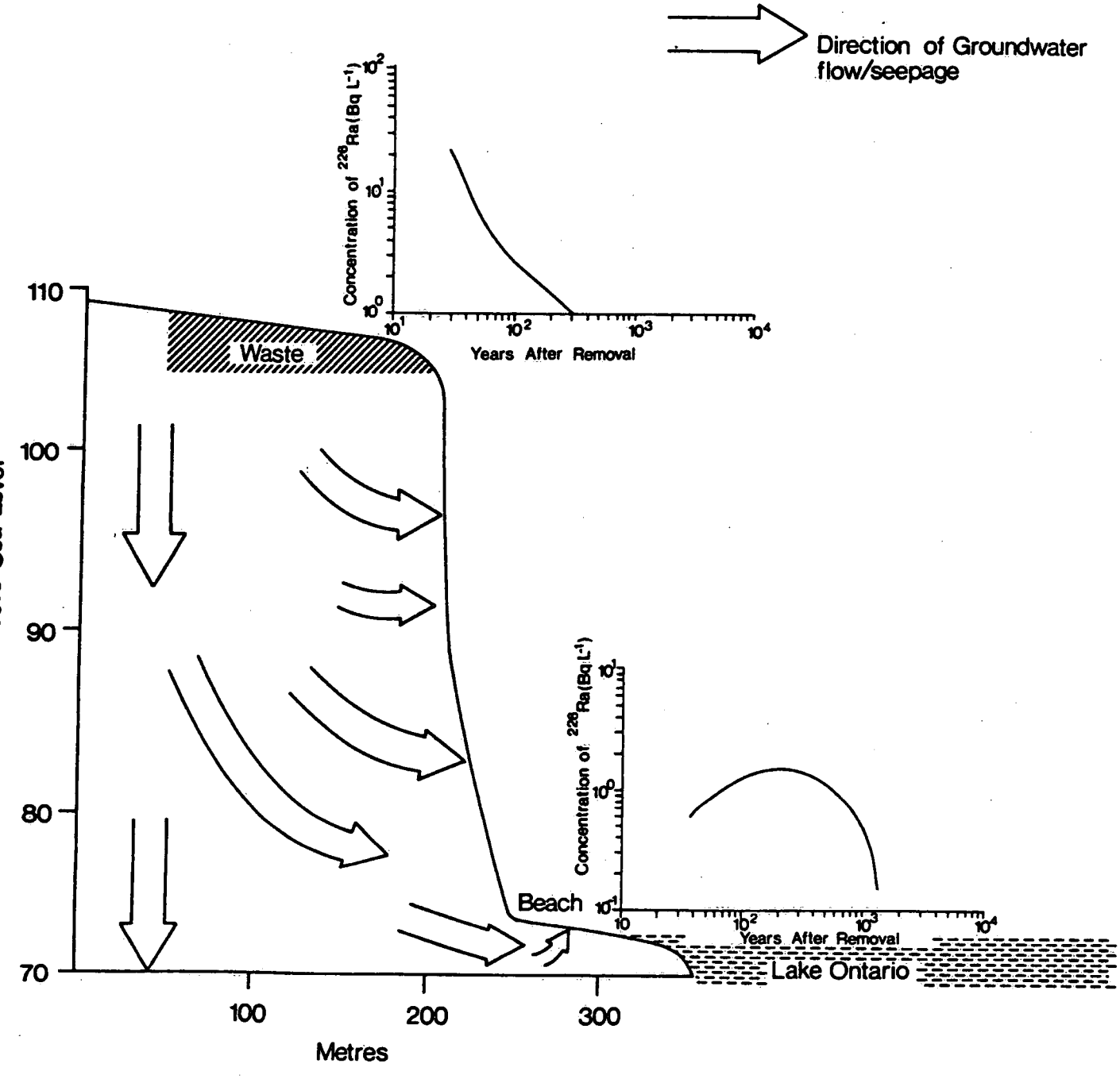
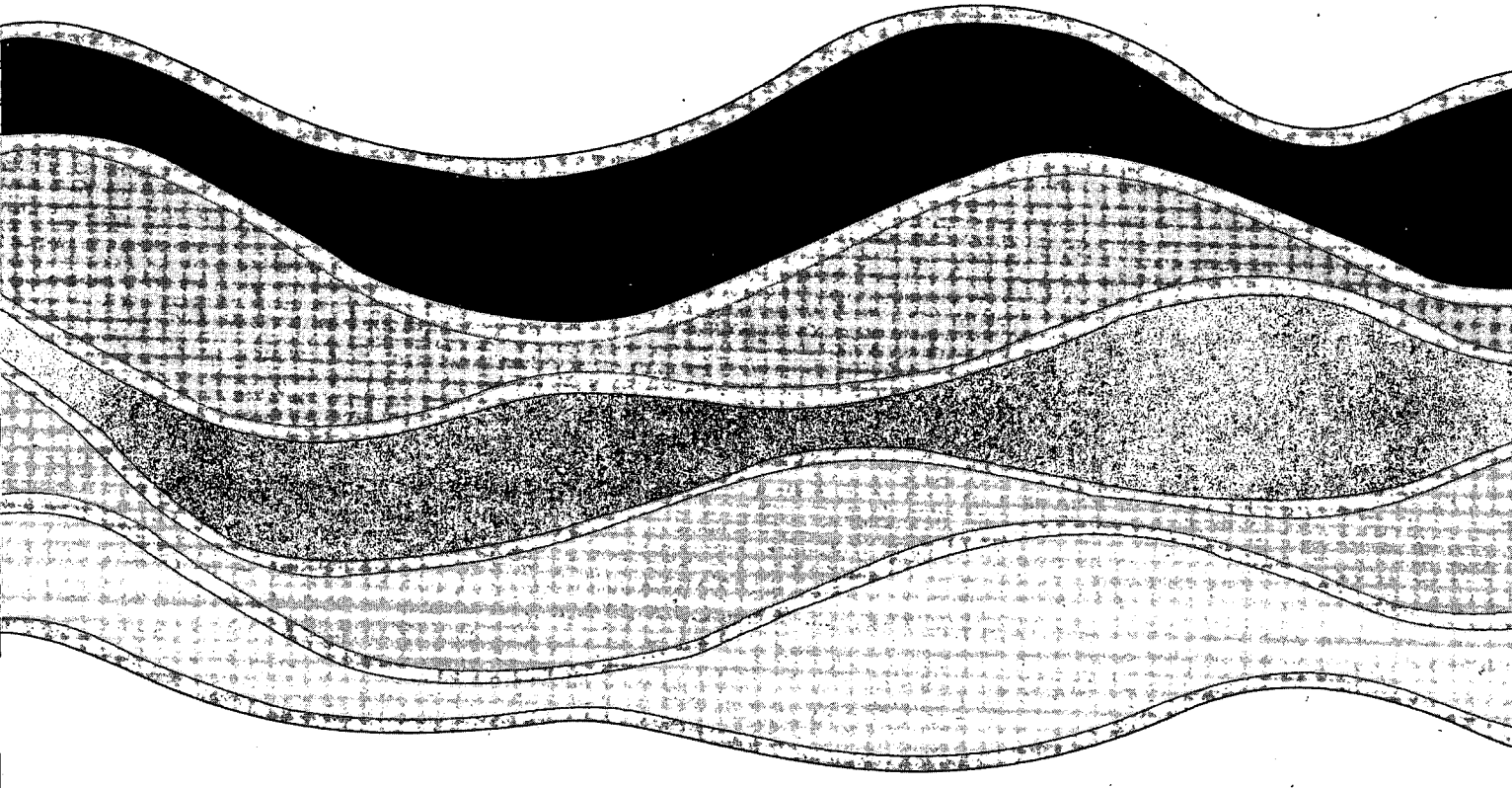

Direction of Groundwater flow/seepage
Concentration of ^{226}Ra (Bq L^{-1})
10^2
10^1
10^0
10^1
10^2
10^3
10^4
Years After Removal
Waste
110
100
90
80
70
Concentration of ^{226}Ra (Bq L^{-1})
10^2
10^0
10^{-1}
10
10^2
10^3
10^4
Years After Removal
Beach
Lake Ontario
100
200
300
Metres

FIGURE 21



NATIONAL WATER RESEARCH INSTITUTE
P.O. BOX 5050, BURLINGTON, ONTARIO L7R 4A6

Environment Canada Environnement Canada

Canada

INSTITUT NATIONAL DE RECHERCHE SUR LES EAUX
C.P. 5050, BURLINGTON (ONTARIO) L7R 4A6