Inventory of Radionuclides for the Great Lakes

Nuclear
Task
Force

International
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INVENTORY OF RADIONUCLIDES FOR THE GREAT LAKES

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OVERVIEW

The Nuclear Task Force of the International Joint Commission, United States and Canada, has prepared a document, "*Inventory of Radionuclides for the Great Lakes*." This report uses a material balance approach to describe the quantities and composition of reported releases of radionuclides to air and water, and the distribution of these nuclides in atmospheric, aquatic and biotic compartments of the ecosystems of the Great Lakes. The material balance approach is a first step in establishing the sources, pathways, distributions and movements of radionuclides for the purposes of making specific assessments of human and ecosystem exposure, and undertaking risk assessments from the exposure information.

The primary anthropogenic source of radionuclides is the discharge from nuclear fuel cycle facilities in the Great Lakes region. The contribution of radioactive debris from the atmospheric testing of nuclear weapons has decayed sufficiently in the thirty five years since the Limited Nuclear Test Ban Treaty (1963) that it is a major source only in areas not currently receiving discharges from nuclear facilities. Other sources which use radionuclides are commercial, industrial, medical and research institutions. These sources use very small quantities of radioactive materials, but the very large number of such sources may make, in the aggregate, a significant contribution to the burden of radioactive materials in the environment.

The Task Force has concluded that monitoring of radionuclides in the Great Lakes primarily meets the need for compliance by users of radioactive materials with the conditions of the licenses for discharge. This results in differences in the radionuclides reported, how radionuclide levels in the environment are reported, the extent of off-site monitoring, and the specific biological compartments included in monitoring by facilities in Canada and the United States. Very little of the monitoring activities are designed to address or are capable of considering the movement and cycling of radionuclides through environmental compartments and ecosystems. Nevertheless, this report begins to consider in a systematic but limited way the cycling of radionuclides through biota. A revised monitoring and analytical protocol with emphasis on biouptake characteristics, physiological roles and impacts would greatly help in meeting the goals and objectives of the Great Lakes Water Quality Agreement of 1978, as amended by the Protocol of 1987.

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1 INTRODUCTION

1.1 Mandate of Task Force

In 1995, the International Joint Commission (hereafter: the Commission or IJC) authorized a "Nuclear Task Force" (hereafter: the Task Force) to review, assess, and report on the state of radioactivity in the Great Lakes and to carry out such other activities as the Commission might, in future, so direct. As an initial project, the Commission requested a review and assessment of the status of radioactivity in the Great Lakes. That project had a completion time within the 1995–1997 biennial Great Lakes reporting cycle. In addition, the Commission authorized the Task Force to recommend additional projects based in part on the following criteria:

(a) Work performed in the preparation of the report on the state of radioactivity in the Great Lakes. The Task Force would use this report as the principal vehicle upon which to base recommendations for various projects. An objective of the report was the prioritization of nuclear problems in the Great Lakes requiring analysis and remediation.

(b) Concerns of the Commissioners.

(c) Problems brought to the attention of the Task Force by its Members, Associates, and others in the course of its work.

The Commission requested that the Task Force make its first official recommendation of additional projects upon the completion of work for its first assessment of the state of radioactivity in the lakes. Further, the Task Force shall undertake such other projects as the Commission directs and shall seek from the Commission directly whatever resources and funds are needed for specific projects and support. This report responds to the mandate set forth by the Commission in 1995.

The Task Force determined that an "Inventory of Radionuclides" for the Great Lakes was an essential project to address the "state of radioactivity" in the Lakes. Following that decision, the Task Force undertook a study to produce such an inventory. This report results from that work.

1.2 Scope of the Inventory

An inventory of radionuclides for the Great Lakes attempts to quantify and organize the information on the sources, levels, distributions, receptors, and repositories of radioactivity in the Great Lakes Basin (hereafter: Basin). It is the numerical part, but not a theoretical modeling simulation, of a material balance of radioactive substances found in the Great Lakes.

The Commission's Great Lakes projects derive from an international agreement between the United States and Canada called the "Great Lakes Water Quality Agreement" (hereafter: the Water Quality Agreement or Agreement; IJC 1987*a*). The Agreement contains a "specific objective" (numerical environmental criterion) for radioactivity. In the 25 years of the Agreement's existence, neither this objective nor the subject of radioactivity itself drew much Commission attention. With the impending decommissioning of nuclear power plants, the growing problems of nuclear waste, and the signing of a Comprehensive Test Ban Treaty on 24 September 1996, posing a plutonium disposal problem, general concerns about the effects of radioactivity on humans and ecosystems have made the subject of radioactivity very timely.

The Agreement also espouses an "ecosystem approach," which the Task Force has used to place in perspective the extent to which radionuclides may be environmental factors in the dynamics of Great Lakes ecosystems. This ecosystem approach requires, where possible, an estimation of the radioactivity stored in Lake biota, sediments, and the water column.

An inventory is a natural starting point to evaluate many of the issues of radioactivity. It basically organizes the information on what exists and where it exists. Without an inventory, the basic analysis of risk assessment cannot be performed, nor can one determine which aspects of sources, distributions, and pathways of radionuclides require special attention. The Commission recognized the fundamental importance of an inventory and authorized the Task Force to develop this tool.

1.3 History

From 1945 to 1963, radioactive fallout from the atmospheric testing of nuclear weapons was the main source of artificial radioactivity to the Great Lakes. Starting in 1963, the commissioning of the reactors at nuclear power plants in the Basin added new sources of artificial radioactivity to the Great Lakes. Following the Limited Test Ban Treaty in 1963, atmospheric testing of nuclear weapons continued sporadically through 1980. Simultaneously, the number of nuclear power plant facilities in the Great Lakes region increased rapidly until 1974 then more slowly until 1993. There are 11 nuclear power plants with 16 reactors in the United States portion of the Basin and 4 nuclear power plants with 21 reactors in the Canadian portion of the Basin, all with emissions to the Basin. Two other nuclear power plants operate in Great Lakes states near the Basin, but their emissions enter other watershed and airshed regions. During the 30-year period of the Limited Test Ban Treaty, the decay of residual nuclear debris from atmospheric testing has reduced nuclear fallout sufficiently to make it a secondary source of artificial radioactivity to the Great Lakes.

Other large source of radioactivity in the Basin include the following: a tritium removal plant at Darlington, uranium mine and mill tailings that enter the Serpent River Region, uranium refining and conversion at Blind River and Port Hope, and weapons facilities and auxiliary operations at Ashtabula. Not all of these facilities are currently operating, but they remain sources of radioactivity to the Basin. There is some question as to whether facilities at Fernald (Ohio) can produce emissions that reach the Basin directly. The Task Force notes that this facility is sufficiently close geographically to the Basin to consider it as a possible source.

Previous Commission reports from 1977 to 1987 (IJC 1977, 1983, 1987*b*) have reviewed radioactivity in the Great Lakes Basin, specifically those of the Great Lakes Water Quality Board (hereafter: GLWQB). These reports discussed the routinely studied radionuclides: tritium (^{3}H), strontium (^{90}Sr), cesium (^{137}Cs), radium (^{226}Ra), uranium (^{238}U), and iodine (^{131}I); total α, β, and γ radiation; and a few occasionally reported radionuclides: antimony (^{125}Sb), cobalt (^{60}Co), and thorium (^{232}Th). The past reports can help to address the amount of the radioactivity in the Great Lakes, but they are an inadequate basis for addressing such issues as ecosystem impacts of radioactivity, the technology and resource needs for nuclear waste isolation, the decommissioning of nuclear reactors, and interactions of toxic chemicals and radiation in the ecosystem.

1.4 Exclusion of direct cosmic and terrestrial radiation

The Task Force did not consider the radiation associated with direct cosmic ray bombardment, except where such radiation produces cosmogenically important background radionuclides. Thus, the Task Force did not consider the problems of solar flux, UV-B radiation, and similar topics as these radiations are non-ionizing and pose a different set of problems from those of radionuclides. Further, the Task Force omitted the natural production of radon gas a decay product of radium in undisturbed rock and soils. Rather, the Task Force addressed the subject of radon gas in its consideration of radionuclides produced in the nuclear fuel cycle, under the topic of uranium mining and milling and fuel processing.

1.5 Acknowledgments

The Task Force used only data from open and public sources, including recently declassified documents from the atomic energy authorities in Canada and the United States. When the Task Force learned that some data were "Proprietary," it sought the permission for use of the data from its holders. The Task Force gratefully acknowledges the providers of data and many persons who assisted in its acquisition and use. In particular, the Task Force thanks Messrs. Paul Gunter and Marcel Buob of the Nuclear Information Resources Service (NIRS) for compiling and organizing the data on nuclear facility licenses, emissions, and monitoring protocols from the files in the reading room of the United States Nuclear Regulatory Commission (NRC); Mr. Michael Petko of the United States Environmental Protection Agency (EPA) for supplying the reports of the Environmental Radiation Monitoring System; Dr. Lester Machta of the National Oceanic and Atmospheric Administration (NOAA) for supplying several special reports from the National Council on Radiation Protection (NCRP); Mr. William Condon of the New York State Department of Health, Bureau of Environmental Radiation Protection, for supplying the annual reports of his organization; Ontario Hydro for permission to access and use proprietary data from its monitoring and surveillance reports and other documents related to nuclear power generation in Ontario; the Atomic Energy Control Board (AECB) of Canada for assistance in obtaining information on the use of radionuclides by hospitals and commercial entities in Ontario; the reference librarians at the AECB library in Ottawa for patiently assisting us with various reference tasks; Dr. Ursula Cowgill for assistance in analysis and understanding of the data on elemental cycling in plants and animals; Dr. David Edgington for assistance in analyzing inventories for transuranic elements; Mr. Paul Payson for his excellent work in editing this report and last, but never least, the indefatigable and dedicated assistance of Ms Émilie Lepoutre, who served as the Task Force's secretary, assistant, telephone operator, and good friend.

1.6 Stylistic Conventions

The stylistic conventions of the Canadian National Research Council's Research Press have been followed as closely as possible in the preparation of this document.

2 INVENTORY BY SOURCE OF RADIOACTIVITY

2.1 Description of Sources Pertinent to the Great Lakes

The sources of radioactivity include natural background radiation from cosmogenic and terrestrial sources, residual debris from weapons testing in fallout, atmospheric deposition of radionuclides emitted in gaseous discharges from various facilities, liquid emissions from various facilities, and many smaller sources that require identification. Facilities include uranium mining and milling operations, refining, processing, fuel fabrication, nuclear power plants, waste management facilities, and reprocessing and tritium-recovery operations. Smaller sources include research nuclear reactors, radiochemical and radiobiological laboratories, hospital nuclear medicine departments, and industrial sources. The various sources are discussed separately, along with comments on the data acquisition, analysis procedures, and reported emissions. The Task Force made no judgements as to the potential hazards implied by the sources.

2.2 Natural Sources of Radioactivity in their Undisturbed State

The Task Force first examined the natural background of radionuclides for the Great Lakes Region. Since the earth's beginning, every component of the environment has been exposed to a natural level of radioactivity. This natural background forms a baseline against which to evaluate the levels and impacts of other sources of radioactivity in the Region, and an inventory of natural radionuclides in this natural background provides clues to the inventories of other radionuclides of the same elements that arise from other processes and sources.

What is the natural background for radiation? Because what some groups have called "natural," other groups have called "technically or technologically enhanced," the Task Force adopted the following definition:

> *The **natural background radiation** levels in the Great Lakes Basin are those derived from cosmic rays and from natural geochemical materials in **undisturbed** strata.*

The Task Force further based its analysis on the earliest reliable measurements it could find. Increases in natural radiation since then belong to the category TENR (technologically enhanced natural radioactivity), which refers to the increase in apparent natural or background radiation resulting from some technical activity (*e.g.*, mining of uranium ore, which leaves radioactive tailings exposed to the atmosphere).

There are two sources of natural background radiation: the interaction of cosmic radiation with various atoms in the atmosphere and a fixed geochemical quantity of naturally occurring radionuclides in the earth's crust. Atmospheric processes include the bombardment of stable nuclei by cosmic rays, other radioactive particles and atomic particles, as well as collisions between selected stable nuclei. Some terrestrial radionuclides decay to gaseous elements (*e.g.*, radon), which reach the earth's surface through diffusion through soil layers and can then enter the atmosphere. The cosmogenic production of radionuclides and release to the atmosphere of radionuclides from crustal processes occur at rates that are balanced by the decay of the radionuclides produced and released. The natural production of atmospheric radionuclides is essentially a steady-state process, provided the cosmic ray flux and the concentration of target atoms remain constant.

Radionuclides ultimately decay to stable (non-radioactive) radionuclides. Further, the decay of radioactive crustal material continuously decreases the natural radioactive content of terrestrial materials. This assures that, over geological time (millions of years), the total inventory of natural background radionuclides declines globally and systematically.

Assessing the natural background of a radionuclide is often a difficult task, and some of the estimates include scientific and political controversies. Although the natural background level should either remain constant or decrease, sometimes a situation arises of an apparent increase in natural background levels reported for a radionuclide, usually explained by either various environmental processes that transport or translocate radionuclides from one region to another (*e.g.*, climate processes, oceanic movements, geophysical upheaval) or that past monitoring of background levels of a given radionuclide was not sufficient by today's standards to quantify the sources of a radionuclide. Once in the environment, contributions from various natural sources (in disturbed and undisturbed states relative to the natural occurrence) and artificial sources were subject to the various mixing processes that incorporate the radionuclide into the ambient observed radiation. This observed ambient radiation is sometimes equated to natural background, although it actually represents some unknown summation of contributions from natural and artificial sources. With regard to past monitoring of background levels, the Task Force notes the importance of knowing whether measurements began after rather than before the onset of nuclear weapons testing programs, and the sensitivity, accuracy and calibration standard of the instrumentation used.

A few radionuclides that arise mainly from artificial sources can also arise from natural processes (*e.g.*, ^{3}H and ^{14}C). For certain minor sources, such as nuclear reactions in extraterrestrial dusts and meteorites, the rates of natural production are virtually zero or so small relative to other processes as to be insignificant. Other naturally occurring radionuclides become environmentally active after some human action. Such radionuclides are "technically enhanced" natural radiation. The United States Environmental Protection Agency (EPA) invented the term in the early 1970's, and it "stuck." Examples include uranium and thorium and their decay products, which enter the environment from mining, milling, ore processing, and the burning of fossil fuels with a high radioactive mineral ash.

2.2.1 Primordial (Terrestrial) Radionuclides

If the half-life of a radionuclide found in geological strata approximates the estimated age of the earth, then the radionuclide is *primordial*; it was presumably present from the time of the earth's beginning. Inventories of primordial radionuclides are essential parts of the natural background level of radioactivity in the environment.

Two classes of radionuclides occur naturally in geological strata: those in the decay series of thorium and uranium and those which do not originate from decay series. Uranium and thorium are natural radioactive elements in various minerals and ores as well as trace contaminants in coal and phosphate-bearing rocks. Geologists and geochemists have intensively studied the mineral deposits of uranium and thorium and have compiled extensive and reliable data on mineral inventories. The non-decay series radionuclides include the well-known ^{40}K (potassium-40) and ^{87}Rb (rubidium-87); radioactive forms of vanadium, cadmium, platinum, cerium, and other rare earth (lanthanide) elements; and one isotope of bismuth. Tables 1a and 1b present the primordial and decay series radionuclides and the decay chains of the latter. Of the non-decay series radionuclides, those of ^{40}K, vanadium (^{50}V), and ^{87}Rb are useful in assessing the inventories of unstudied radionuclides.

Several uranium and thorium isotopes can also undergo a spontaneous fission process. This mode of decay is very rare compared with the normal decay mode of alpha disintegration, making it relatively unimportant for purposes of calculating an inventory.

2.2.2 Mobility and Transport of Terrestrial Radionuclides

The atmospheric release of radionuclides from geological strata is a multistage process: formation of a radionuclide of a gaseous element, diffusion of the radionuclide through soils to the soil surface (soil–atmosphere interface), and release to the atmosphere. The most important gaseous radionuclide is the noble gas radon, ^{222}Rn, and its long-lived progeny ^{210}Pb and ^{210}Po. Radon gas has a half-life of 3.8 days. Once radon reaches the atmosphere, it dissipates quickly while continuing its radioactive decay. Its decay products are solids and aerosol-forming radionuclides, which can deposit on soil or water, but inventories of radon *per se* are not important for contamination of the water in the Great Lakes, although radon as an air pollutant within the Great Lakes region may be important in health assessments of residents.

TABLE 1a
PRIMORDIAL RADIONUCLIDES OF TERRESTRIAL ORIGINS

Radionuclide	Half-life (years)	Isotope abundance (%)	Element abundance in crustal rock (ppm)**	Concentration in crustal rock (Bq/kg)
Non series decay				
^{40}K	1.26×10^{9}	0.018	2090	603
^{50}V	6×10^{15}	0.25	135	1×10^{-5}
^{87}Rb	4.8×10^{10}	27.85	90	70
^{113}Cd	1.3×10^{15}	12.26	0.2	7×10^{-7}
^{115}In	6×10^{14}	95.77	0.1	2×10^{-5}
^{123}Te	1.2×10^{23}	0.87	0.002	1×10^{-7}
^{138}La*	1.12×10^{11}	0.089	30	0.02
^{142}Ce*	5×10^{16}	11.07	60	1×10^{-5}
^{144}Nd	1.4×10^{15}	23.87	28	3×10^{-6}
^{147}Sm*	1.05×10^{11}	15.07	6	0.7
^{152}Gd*	1.1×10^{14}	0.20	5.4	7×10^{-6}
^{174}Hf	2×10^{15}	00.163	3	2×10^{-7}
^{176}Lu*	2.2×10^{10}	2.6	0.5	0.04
^{187}Re	4.3×10^{10}	62.93	0.001	1×10^{-3}
^{190}Pt	6.9×10^{11}	00.0127	0.005	7×10^{-8}
^{192}Pt	1×10^{15}	00.78	0.005	3×10^{-6}
^{209}Bi	2×10^{18}	100	0.17	1×10^{-10}
Decay series				
^{232}Th	1.4×10^{10}	100		
^{235}U	7.1×10^{8}	00.72		
^{238}U	4.5×10^{9}	99.27		

* Denotes a lanthanide or rare earth element

** Elemental abundance in crustal rock is the weight abundance of the element in crustal material multiplied by the per cent abundance of the primordial isotope relative to all isotopes of the element. Data are from Mason (1965).

TABLE 1b
THE DECAY CHAINS OF URANIUM AND THORIUM

^{238}U, ^{234}U

$^{238}\text{U} \rightarrow {}^{234}\text{Th} \rightarrow {}^{234}\text{Pa} \rightarrow {}^{234}\text{U} \rightarrow {}^{230}\text{Th} \rightarrow {}^{226}\text{Ra} \rightarrow {}^{222}\text{Rn} \rightarrow {}^{218}\text{Po} \rightarrow {}^{214}\text{Pb} \rightarrow {}^{214}\text{Bi} \rightarrow {}^{214}\text{Po} \rightarrow {}^{210}\text{Pb} \rightarrow {}^{210}\text{Bi} \rightarrow {}^{206}\text{Tl} \rightarrow {}^{206}\text{Pb}$ (stable)

^{235}U

$^{235}\text{U} \rightarrow {}^{231}\text{Th} \rightarrow {}^{227}\text{Ac} \rightarrow {}^{223}\text{Fr} \rightarrow {}^{219}\text{Ac} \rightarrow {}^{215}\text{Bi} \rightarrow {}^{211}\text{Pa} \rightarrow {}^{211}\text{Bi} \rightarrow {}^{207}\text{Tl} \rightarrow {}^{207}\text{Pb}$ (stable)

^{232}Th

$^{232}\text{Th} \rightarrow {}^{228}\text{Ra} \rightarrow {}^{288}\text{Ac} \rightarrow {}^{228}\text{Th} \rightarrow {}^{224}\text{Ra} \rightarrow {}^{220}\text{Rn} \rightarrow {}^{216}\text{Po} \rightarrow {}^{212}\text{Pb} \rightarrow {}^{212}\text{Bi} \rightarrow {}^{212}\text{Po} \rightarrow {}^{208}\text{Tl} \rightarrow {}^{208}\text{Pb}$ (stable)
↓
$^{208}\text{Tl} \rightarrow {}^{208}\text{Pb}$ (stable)

The various isotopes of uranium decay either to radioactive thorium or radioactive neptunium. The most common isotope of uranium, ^{238}U, decays to ^{234}Th. The various isotopes of thorium decay to radioactive isotopes of radium, the most important of which is ^{226}Ra, a decay product of ^{230}Th. ^{226}Ra decays to ^{222}Rn, the only gas in the decay series. The radioactive decay products or radon are radioactive isotopes of lead, bismuth, and polonium, until the chain ends with stable isotopes of lead (Faw and Shultis 1993).

Soil radioactivity includes ^{40}K, the thorium and uranium decay series, and the other natural radionuclides in trace amounts. In particular, radionuclides occur in coal, phosphate-bearing rocks, and extractable minerals. The radioactivity can be released in burning of fossil fuel, building materials, or use of phosphate fertilizers. The GLWQB reports present the data for these source of radioactivity in the Basin (IJC 1977, 1983, 1987*b*). Only the uranium mine and mill tailings have been included as important point sources of radioactivity in the inventory of radionuclides in the Basin. Radionuclides in fertilizers should be considered under non-point-source pollutants. Building materials may pose local problems but do not appear to be an important source of radioactivity to the Basin. The Task Force considers that the radioactivity in fossil fuel emissions remain an unquantified source of radioactivity.

2.2.3 Cosmogenically Formed Radionuclides

The interaction of cosmic radiation with the earth's atmosphere produces many radionuclides. At high altitudes, hydrogen atom nuclei (protons) are 95% of the available targets subject to cosmic ray bombardment. Other atmospheric targets include ions and nuclei of helium, argon, krypton, oxygen, and nitrogen and the carbon atoms in carbon dioxide and carbon monoxide. Many cosmogenically produced radionuclides have short half-lives and do not affect the inventory of natural radionuclides. Some can serve as tracers for small-scale atmospheric processes (Reiter 1975). Table 2 lists the major cosmogenically produced radionuclides.

TABLE 2
COSMOGENICALLY PRODUCED RADIONUCLIDES

(a) Major radionuclides of importance to the Great Lakes			
^{3}H	^{7}Be, ^{10}Be	^{14}C	^{32}P, ^{33}P
^{41}Ar	^{81}Kr		
(b) Other radionuclides of scientific interest			
^{11}C*	^{13}N	^{22}Na, ^{24}Na	^{26}Al, ^{28}Al*
^{28}Mg*	^{35}S, ^{38}S	^{37}Ar, ^{39}Ar	^{18}F*
^{31}Si*, ^{32}Si	^{41}K*, ^{42}K*	^{43}Sc*, ^{44}Sc*	^{49}Ca*
^{56}Mn	^{64}Cu	^{86}Rb	^{34m*}Cl, ^{36}Cl, ^{38}Cl*, ^{39}Cl*

* Denotes the radionuclides that are produced by cosmic ray bombardment only. The others are produced by anthropogenic as well as natural sources.

Tritium (^{3}H)

Although tritium is produced in the atmosphere, it is more difficult to determine its natural background, because environmental measurements of tritium began after the onset of nuclear weapons testing. UNSCEAR (1982) reviewed data that suggested that the natural concentration of tritium in lakes, rivers, and potable waters was 0.2–1.0 Bq/L (5–25 pCi/L) prior to the advent of weapons testing.

Most cosmogenically formed tritium deposits in oceans. The small fraction that goes to the Great Lakes can be estimated by comparing the size of the Great Lakes with that of the oceans. Table 3 summarizes data on the inventory of tritium of cosmogenic origin.

Beryllium-7,10

Two radioactive beryllium radionuclides, ^{7}Be (half-life: 53.6 days) and ^{10}Be (half-life: 2.6 million years), are produced cosmogenically, mainly in the stratosphere. Exchanges between atmospheric compartments produce a slow build-up of these isotopes in the troposphere. Production of ^{7}Be occurs mainly at higher latitudes and shows a seasonal variation in rainfall with maximum values in spring of about 4 mBq/m^{3} and minimum values of about 1.5 mBq/m^{3} (Kolb 1970).

Beryllium deposited in wet and dry fallout goes mainly to sediments and terrestrial soils. Land surfaces accumulate 71% of ^{7}Be, and aquatic surfaces receive 28%. Deep ocean sediments receive 71% of the ^{10}Be, and terrestrial areas receive 28%. The differences in the inventories for the two isotopes reflects the differences in their half-lives: the longer lasting ^{10}Be reaches terrestrial and aquatic repositories before it has decayed significantly. According to the National Council on Radiation Protection and Measurements (1975), the depositional rates of the two beryllium isotopes are relatively constant. Table 4 presents the beryllium data.

Carbon-14

Probably more is known about the natural background of ^{14}C than any other cosmogenically produced radionuclide. ^{14}C is produced by neutron bombardment of ^{14}N in the atmosphere. The variations in the atmospheric content of ^{14}C are caused by changes in the cosmic ray flux. The neutron bombardment of ^{14}N also follows airborne detonation of nuclear weapons. Libby (1958) estimated that each equivalent ton of TNT explosive produced an average of 3.2×10^{26} atoms of ^{14}C. [A gram-atom (14 grams) of ^{14}C contains 6.023×10^{23} atoms, and each ton of equivalent TNT explosive in a nuclear weapon produces approximately 7,500 grams of ^{14}C.] ^{14}C rapidly oxidizes to carbon dioxide (CO_2), and moves environmentally in this form. Because oceans receive most of the carbon dioxide, understanding the behavior of ^{14}C requires a model of global transport with atmosphere–ocean coupling processes.

In discussing ^{14}C relative to ^{12}C, researchers sometimes use the terms "normal ratios" and "excess ratios." The *normal ratio* refers to the ratio of $^{14}C/^{12}C$ that results from the natural production of these two isotopes. When the ratio of radioactive to stable carbon found in some sample exceeds the normal ratio, that portion of the ratio unaccounted for by natural processes is called *excess*. Occasionally a researcher reports a reference value of the geochemical ratio, $^{14}C/^{12}C$, calls it " normal," and then designates any observed increase in the ratio over his/her value as an excess ratio for certain analysis. Such reference values used in technical papers need to be checked against normal ratios.

UNSCEAR (1977) noted that "the fossil records of ^{14}C in tree rings and lake and ocean sediments suggest that the natural ^{14}C levels have remained relatively unchanged for many thousands of years. ... the long-term fluctuation over a period of 10,000 years is attributed to a cyclical change of the dipole strength of the earth's magnetic field, which results in a cyclical change of the cosmic ray flux, which in turn changes the ^{14}C production rate." UNSCEAR thus implied that the normal geochemical ratio value of $^{14}C/^{12}C$ has been constant since primordial times despite different estimates for the natural production rate of radiocarbon, ranging from a low of 1.8 $atoms \cdot cm^{-2} \cdot s^{-1}$ to a high of 2.5 $atoms \cdot cm^{-2} \cdot s^{-1}$. UNSCEAR cited as a "currently most accepted" value, 2.28 $atoms \cdot cm^{-2} \cdot s^{-1}$, although some geochemists have long used the upper value of 2.5 $atoms \cdot cm^{-2} \cdot s^{-1}$ in calculations of a radiocarbon inventory.

TABLE 3
INVENTORIES OF COSMOGENICALLY PRODUCED TRITIUM (^3H)

Half-life	12.33 years
Production Rate (atoms·cm^{-2}·s^{-1})	
Troposphere	8.4×10^{-2}
Total Atmosphere	0.25
Global Inventory	3.5 kg (1300 PBq) (as of 1967)
Distribution (%)	
Stratosphere	6.8
Troposphere	0.40
Biosphere/land surface	27.0
Mixed ocean layer	35.0
Deep ocean layer	30.0
Ocean sediments	0
Concentration	
Stratosphere	0.10 Bq/kg air
Troposphere	0.00012 Bq/kg air
Oceans	0.00067 Bq/kg water (average)
Oceans	0.11 Bq/kg water (surface water)
Method of Production	Spallation processes; Target nuclei ^{14}N and ^{16}O
Main Radiation	β 18.6 keV

Other Reported Global Inventory Calculations

Atmospheric Production Rate (atoms·cm^{-2}·s^{-1})	Inventory (PBq)
0.10–0.20	500–1000 (as of 1953)
0.12	600 (as of 1954)
0.14	700 (as of 1955)
1.2	6300 (as of 1957)
1.06	5500 (as of 1958)
0.9	4800 (as of 1958)
0.75	3700 (as of 1958)
0.6–1.3	3000–7000 (as of 1961)
0.25–0.35	1300–5500 (as of 1962)

Notes: (1) Inventory per Teegarden (1967).
(2) Multiple inventory estimates for the same year indicate different investigators.

TABLE 4
INVENTORIES OF COSMOGENICALLY PRODUCED BERYLLIUM (^{7}Be and ^{10}Be)

	^{7}Be	^{10}Be
Half-life	53.6 days	2.5×10^{6} years
Production Rate (atoms·cm^{-2}·s^{-1})		
Troposphere	2.7×10^{-2}	1.5×10^{-2}
Total Atmosphere	8.1×10^{-2}	4.5×10^{-2}
Global Inventory	3.2 kg (107 PBq)	3.9×10^{5} kg (as of 1967)
Distribution (%)		
Stratosphere	60.0	0.00037
Troposphere	11.0	0.23
Biosphere/land surface	8.0	29.0
Mixed ocean layer	20.0	0.00057
Deep ocean layer	0.2	0.001
Ocean sediments	0	70.7
Concentration		
Stratosphere	0.28 Bq/kg air	—
Troposphere	0.011 Bq/kg air	—
Oceans	—	2×10^{-7} Bq/kg water
Method of Production	Spallation processes; Target nuclei ^{14}N, ^{16}O, ^{12}C	
Main Radiation	Electron capture γ 477 keV	β 555 keV

Table 5a presents the inventory data for ^{14}C based on information available before 1970. In 1972, a revised estimate of the average rate of production of the radionuclide in the atmosphere over the 11-year solar cycle suggested a slightly lower value than given in Table 5a. Also, UNSCEAR (1977) presented a method of estimating a natural inventory based on "units of atmospheric content of^{12}C" (referred to herein as "carbon units"), the main stable isotope of carbon. Those additional inventories appear in Table 5b.

The "carbon units of ^{12}C" are multiples of 6.17×10^{17}, which Broecker et al. (1960) used as an estimate of the stable carbon atom content of the atmosphere in 1963. This estimate would suggest that the biosphere (atmosphere and oceans) contains 67 units of carbon. However, Broecker reported that the concentrations of ^{14}C in carbon units in the surface ocean and deep ocean were lower than concentrations of the isotope in the atmosphere by 4 and 17%, respectively. This would result in inventories of ^{14}C in the combined surface ocean and deep ocean compartments of 56 units of the activity of^{14}C (about 3.8 MCi), and revise the atmospheric production to 2.28 atoms·cm^{-2}·s^{-1}. Broecker then estimated that about 8% of the inventory of ^{14}C was in oceanic sediments, which corresponds to a total inventory for natural production of ^{14}C of about 230 Mci.

Krypton-81 and Argon-37, 39, 41

Cosmogenic processes produce several radionuclides of noble gases: ^{81}Kr and $^{37, 39, 41}$Ar. The inventories for these radionuclides are mainly limited to the atmosphere. Only ^{81}Kr decays to stable krypton. Argon radionuclides decay to isotopes of potassium and chlorine, which combine rapidly with oxygen and water vapor to form oxides and hydroxides. These attach to particulate matter and deposit on terrestrial and aquatic media. Table 6a presents selected data for two of these noble gas radionuclides.

Radionuclides Formed by Neutron Bombardment of Argon Nuclei

Several important radionuclides of atomic weights 20–50 form cosmogenically by spallation reactions of neutron and other particle bombardment of stable argon nuclei. These include such long-lived radionuclides as chlorine-36 (^{36}Cl, half-life: 3.08 × 10^5 years), aluminum-28 (^{28}Al, half-life: 7,400 years), and silicon-32 (^{32}Si, half-life: 280 years) as well as shorter lived radionuclides such as phosphorus-32 and -33 . Table 6b presents selected data for several of these radionuclides.

TABLE 5a
BACKGROUND INVENTORY OF COSMOGENICALLY PRODUCED CARBON (^{14}C)

Half-life	5730 years	
Production Rate (atoms·cm^{-2}·s^{-1})		
Troposphere	1.1	
Total Atmosphere	2.5	1.5 Bq/year
Global Inventory	6.8 × 10^4 kg	1100 PBq (as of 1967)
Distribution (%)		
Stratosphere	0.30	
Troposphere	1.6	
Biosphere/land surface	4.0	
Mixed ocean layer	2.2	
Deep ocean layer	92.0	
Ocean sediments	0.4	
Concentration		
Stratosphere	— Bq/kg air	
Troposphere	— Bq/kg air 0.0048Bq/kg	
Oceans	water	
Method of Production	Spallation processes; Target nuclei ^{14}N and ^{16}O	
Main Radiation	β 166keV	

TABLE 5b
COSMOGENICALLY PRODUCED CARBON (^{14}C) AS SUGGESTED BY BROECKER (1963) AND UNSCEAR (1977)

Production Rate (atoms·cm^{-2}·s^{-1})		
Troposphere	1.1	
Total Atmosphere	2.28	1.4 Bq/year
Global Inventory	5.21 × 10^4 kg	8500 PBq
Distribution (%)		
Stratosphere	0.48	
Troposphere	1.2	
Biosphere/land surface	1.6	
Mixed ocean layer	1.4	
Deep ocean layer	87.3	
Ocean sediments	8.0	

TABLE 6a
INVENTORIES OF COSMOGENICALLY PRODUCED ^{81}Kr AND ^{41}Ar

	^{81}Kr	^{41}Ar
Half-life	21 × 10^5 years	1.83 hours
Production Rate (atoms·cm^{-2}·s^{-1})		
Troposphere	—	—
Total Atmosphere	1.5 × 10^{-7}	45 × 10^{-4}
Global Inventory	16.2 kg	0.014 g (23 PBq)
Distribution (%)		
Stratosphere	16.0	100
Troposphere	82.0	0
Biosphere/land surface	0	0
Mixed ocean layer	0.04	0
Deep ocean layer	0.02	0
Ocean sediments	0	0
Concentration		
Stratosphere	—	
Troposphere	—	
Oceans	—	
Method of Production	Cosmic ray bombardment of stable Kr nuclei	Cosmic ray bombardment of stable Ar nuclei
Main Radiation	X-ray	β 1.196 MeV

Notes: (1) Global inventory data as of 1967.

TABLE 6b
RADIONUCLIDES PRODUCED COSMOGENICALLY BY NEUTRON BOMBARDMENT OF ARGON — PART I: CHLORINE (^{36}Cl), SULFUR (^{35}S), AND ALUMINUM (^{26}Al)

	^{36}Cl	^{35}S	^{26}Al
Half-life	3.08×10^{5} years	87.9 days	7.4×10^{5} years
Production Rate (atoms·cm^{-2}·s^{-1})			
Troposphere	4×10^{-4}	4.9×10^{-4}	3.8×10^{-5}
Total Atmosphere	1.1×10^{-3}	1.4×10^{-4}	1.4×10^{-5}
Global Inventory	1.4×10^{4} kg	4.5 g (1 PBq)	1000 kg
Distribution (%)			
Stratosphere	0.0001	57.0	0.00013
Troposphere	0.8	0.000006	0.0000007
Biosphere/land surface	10.0	29.0	29.0
Mixed ocean layer	24.0	0.014	0.0014
Deep ocean layer	0.4	69.0	0.007
Ocean sediments	0	0	0.007
Concentration			
Stratosphere	0.0048 Bq/kg air	—	—
Troposphere	0.00013 Bq/kg air	—	—
Oceans	—	0.28×10^{-3}	0.6×10^{-3}
Main Radiation	β 714 keV	β 167 keV γ 1.88 MeV γ 1.6, 2.17 MeV from ^{38}Cl daughter isotope	ß(+)1.17 MeV γ 1.81 MeV γ 511 keV

Notes: (1) Inventory as of 1967.

TABLE 6b (continued)
RADIONUCLIDES PRODUCED COSMOGENICALLY BY NEUTRON BOMBARDMENT OF ARGON — PART II: SODIUM (^{22}Na), PHOSPHORUS (^{32}P AND ^{33}P), AND SILICON(^{32}Si)

	^{22}Na	^{32}P	^{33}P	^{26}Si
Half-life	2.62 years	14.28 days	24.4 days	280 years
Production Rate (atoms·cm^{-2}·s^{-1})				
Troposphere	2.4×10^{-5}	2.7×10^{-4}	2.2×10^{-4}	5.4×10^{-5}
Total Atmosphere	1.8×10^{-5}	8.1×10^{-4}	6.8×10^{-4}	1.6×10^{-4}
Global Inventory	1.9–2.0 kg	0.4 g	0.6 g (3PBq)	1.4 kg
Distribution (%)				
Stratosphere	25.0	60.0	64.0	0.19
Troposphere	1.7	24.0	16.0	0.011
Biosphere/land surface	21.0	4.7	5.6	29.0
Mixed ocean layer	44.0	13.0	11.0	0.35
Deep ocean layer	8.0	7.0	1.0	68.0
Ocean sediments	0	0	0	2.8
Concentration				
Stratosphere	8.5×10^{-5} Bq/kg air	0.028 Bq/kg air	0.025 Bq/kg air	—
Troposphere	1×10^{-7} Bq/kg air	1×10^{-4} Bq/kg air	1×10^{-4} Bq/kg air	—
Oceans	—	—	—	4.66×10^{-7} Bq/kg water
Main Radiation	β+ 0.545 MeV β+ 1.82 MeV γ 1.275 MeV γ 511 keV	β 1.710 MeV	β 248 keV	β 210 keV

Notes:

(1) Inventory as of 1967.

(2) Some distribution data total to greater than 100% because of rounding errors.

2.3 Anthropogenic Sources of Radiation

Anthropogenic sources refer to those that are mainly human in origin: military, industrial, educational, recreational, medical, or somehow reflecting a human use, intervention, or process. The two main anthropogenic sources are the fallout of military weapons testing and the generation of electrical power at nuclear power plants. Medical, commercial, and other sources are many in number, but their emissions are individually very small, raising the possibility that the sources may, in the aggregate, be a major contributor to the anthropogenic inputs of radioactivity to the Basin.

2.3.1 Fallout from Atmospheric Testing of Nuclear Weapons

Nuclear technologies over the past 50 years have introduced significant quantities of artificial radionuclides into the global environment. Historically, the greatest part of this radioactivity has come from atmospheric nuclear weapons tests conducted prior to the 1963 Limited Test Ban Treaty, although tests were carried out since then by non-signatory nations. Fallout from the tests has been distributed globally, with the maximum occurring in the North Temperate Zone, which encompasses the Great Lakes Basin. From 1963 to 1996, many weapons tests were carried out underground. Radioactive material occasionally vented to the atmosphere from these tests, but the impact on global fallout was minimal (UNSCEAR 1993). With the signing of the Comprehensive Test Ban Treaty in 1996, even this source of radioactivity has hopefully been eliminated.

Previous Commission reports (IJC 1977, 1983, 1987) have extensively covered the inputs of radionuclides from this source . Since 1987, atmospheric inputs have not been significant. The Task Force briefly reviews this topic to complete the inventory of radionuclides currently stored in the water column and sediments of the Great Lakes. It is recognized, however, that radioactive fallout deposited on land will also eventually make its way into waters through weathering, surface runoff, ground water movement, and various mechanisms of biological incorporation and biological decay.

Of the many radionuclides produced by nuclear detonations, ^{3}H, ^{14}C, ^{90}Sr, and ^{137}Cs have received the greatest attention in environmental monitoring programs. They have been measured in air, water, soil, and food products. Other important radionuclides include ^{95}Zr, ^{95}Nb, ^{106}Ru, ^{131}I, ^{144}Ce, ^{239,240}Pu, ^{241}Pu, and ^{241}Am. Most of radionuclides listed, except the plutonium and americium isotopes, emit beta radiation. Plutonium and americium emit alpha radiation. Although many of the individual radionuclides mentioned are not monitored, agencies of both countries typically report measurements of gross beta radiation. After the brief rise in 1986 due to the Chernobyl accident, the radioactive fallout in the Basin is approaching a level that is entirely due to naturally occurring radionuclides, especially ^{210}Pb. The decreasing trends for the Great Lakes Basin are similar to values across Canada and the United States. The total inventories of the most important fallout radionuclides are reported in the section on **Radionuclide Inventory of the Great Lakes.**

The lifetime or cumulative radiation dose that will be received by individuals in the North Temperate Zone from all atmospheric detonations conducted between 1945 and 1980 is estimated to be about 1.9 mSv (UNSCEAR 1993). In addition to this dose, there will be a small contribution from weapons-generated ^{14}C extending far into the future. Only about 5% of this dose will have been delivered by 2045.

2.3.2 The Nuclear Fuel Cycle Support Industries

The nuclear fuel cycle is currently the main source of anthropogenic radioactivity emitted to the Great Lakes. The cycle consists of mining and milling of uranium; converting the mined uranium to fuel material (typically an oxide of uranium with possible enrichment in^{235}U); fabricating fuel elements (uranium pellets encapsulated into metallic fuel rods); incorporating fuel elements into a nuclear reactor; bringing the reactor to criticality and "burning" of the fuel; reprocessing spent fuel to extract radionuclides for further use; transporting material between fuel-cycle installations; and management of radioactive wastes from each step. All components of the nuclear fuel cycle have been operative in the Great Lakes Basin for some interval of time over the past 35 years.

Uranium Mining and Milling

All uranium mining and milling operations in the Great Lakes Basin are located in the Elliot Lake and Bancroft areas in Ontario.

Elliot Lake once had as many as 15 uranium mining and milling operations. The radioactive tailings of these operations were disposed of in various holding ponds, which empty into the Serpent River and, from it, into Lake Huron. In 1983, the Commission reported that eight mining and milling facilities were operational, and two had closed. By 1987, the Commission had reported that four mines were operational, and one was "under care and maintenance." Further, the Commission noted that there "were several idle and unlicenced tailings areas in the region," which were sources of radionuclides to the environment from leachates. The last operating mine was closed in 1996. The AECB has developed plans for the major waste management areas. These plans feature "the wet cover option" because of acid generation on the mine tailings.

Uranium rock contains, as previously noted, all of the radioactive elements of the uranium and thorium decay chains. The uranium ore from the Elliot Lake area is considered low grade, containing 0.2% natural uranium and 0.4% thorium. Thus, one metric tonne of this ore contains about 2 kg of uranium oxide and has an activity of about 21 MBq from each of the 14 principal members of the^{238}U chain, or a total of about 0.29 GBq. In contrast, the ore in the more recent uranium mining operations in Saskatchewan are high grade and typically contain 2–4% natural uranium. According to UNSCEAR (1982), releases from the mine are primarily radon gas, but the milling operations results in the accumulation of large quantities of tailings containing significant quantities of the uranium decay series isotopes. About 14% of the total radioactivity in the ore feed appears in the uranium concentrate, which achieves better than 90% extraction. According to Ahier and Tracy (1995), Elliot Lake mills extract 95% of the uranium and 10–15% of the other radioactivity. This implies that about 86% of the radioactivity from the uranium decay chain, 0.25 GBq per tonne of ore, and 5% of the uranium, will be retained in the waste as a long-term source of environmental pollution. The principle radionuclides in the waste are radium, thorium, and radon.

Only a very small portion of the radium in uranium ore is water soluble. Mill liquid effluents will vary in activity but will contain all of the uranium decay series radionuclides. The addition of barium chloride ($BaCl_2$) to the holding ponds usually precipitates ^{226}Ra. Table 7 has information on the radionuclides in milling effluents typical of Elliot Lake Mines.

TABLE 7
EFFLUENTS FROM URANIUM MINING AND MILLING

Basis: 1 metric ton of uranium ore yields 2 kg yellow cake

Activity: 20 MBq for each member of ^{238}U decay chain, or total of approximately 300 MBq

Radium in mill liquid effluents: 10–20 Bq/L (prior to treatment)

Treated effluent prior to discharge: 0.3–3.0 Bq/L

Nuclides in liquid effluents prior to any neutralization of acid solutions and precipitation of thorium salts:

^{223}Ra, ^{226}Ra, ^{227}Th, ^{230}Th, ^{232}Th, ^{222}Rn, ^{227}Ac, ^{210}Pb, ^{210}Po

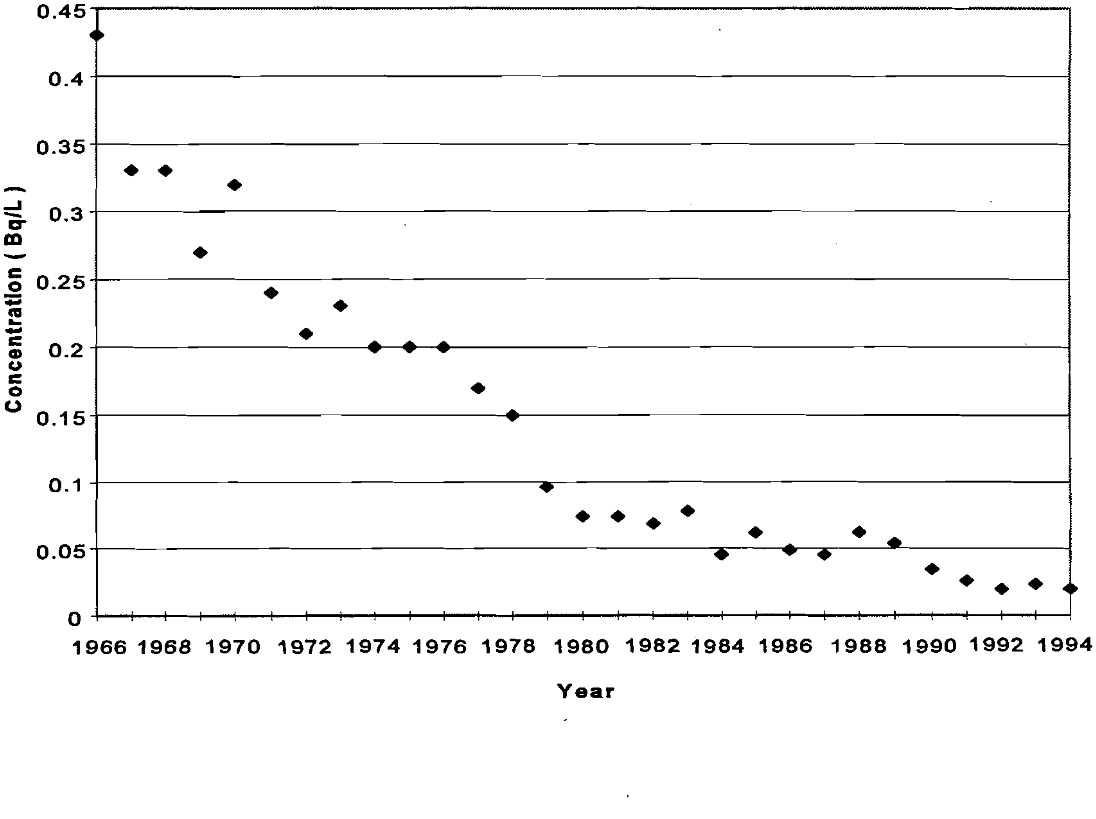
Waste generation (1955–1985): 1.2 × 10^8 m^3 waste rock and tailings (Elliot Lake region)
Represents an activity of 2000 terabecquerels (TBq) of ^{226}Ra (MCi) in 100 megatons (metric) of waste rock, at a concentration of some 18.5 Bq/g. Radon is estimated to emanate from these piles at a rate of 22 $Bq{\cdot}m^{-2}{\cdot}s^{-1}$.

Radon gas originates from the in situ decay of ^{226}Ra. Most radon released to the atmosphere from tailings piles originates in the top surface layers of tailings. Radon from deeper soil layers, the concentrated cake or the decay of radionuclides in rock mass, must diffuse through dense materials, a long process. The readily observed radon production probably occurs mainly in the top 1–2 m of the tailings, although increasing the depth a few more metres does not necessarily change release rates. By covering a tailings pile with clean earth fill and revegetating a tailings area, one reduces the radon release rate by a factor of about 2 for each metre of cover (UNSCEAR 1977). The control of radon emissions is an obvious requirement. Soil or water covers over decommissioned mill tailings are necessary for the control of radon.

Radium in liquid effluents from mining and milling activities is a reported chemical parameter in the monitoring activities under existing permits, but radon is not. Most material reviewed by the Task Force on radon inventories reported estimates based on the authors choosing “typical values” for the airborne release rates for radon and parameters of atmospheric dispersion. The Task Force did not attempt an inventory for radon because of questions about what is a “typical airborne release” and no indications of consistent monitoring of radon.

All of the Elliot Lake mine tailings areas are located within the Serpent River watershed. The Ontario Ministry of the Environment and Energy monitors surface water quality in the Serpent River. One of the monitoring locations (Highway 17) is located downstream of all the mines and provides a good indicator of trends in discharges entering Lake Huron Average annual concentrations of ^{226}Ra at this location are presented in Figure 1.

FIGURE 1. ANNUAL AVERAGE CONCENTRATION OF ^{226}Ra IN THE SERPENT RIVER SURFACE WATER



Uranium Refining, Conversion, and Fuel Fabrication

All uranium fuel fabrication and conversion facilities in the Great Lakes Basin are located in Canada

Most fuel fabrication and conversion facilities in the Great Lakes Basin are in Canada. CAMECO operates facilities at Port Hope and Blind River. Up until 1983 the Port Hope facility consisted of a uranium oxide refinery and a uranium hexafluoride production facility. Then CAMECO moved the refinery operation to Blind River but retained and expanded its conversion facility in Port Hope. The closing of the Port Hope refinery ended the release of ^{226}Ra to Port Hope Harbour. Joshi (1991) noted, however, that the sediments still contain heavy loadings of radionuclides from earlier radium processing (1933–1953) and uranium recovery (1942–1983). Figures 2 and 3 present liquid and airborne emissions respectively for the Port Hope facility, while Figures 4 and 5 show emissions for the Blind River facility.

CAMECO also maintains two low-level radioactive waste management facilities in the vicinity of the Port Hope conversion facility. The Welcome waste management area received waste from the Port Hope uranium processing facility between 1948 and 1955. Although inactive, the site is licensed by the AECB. Groundwater and surface water is collected and treated prior to its release to Lake Ontario. Liquid effluent releases from this site are shown in Table 8.

Similarly the Port Granby waste management area operated from 1955 to 1988. Treated liquid effluent from this facility is shown in Table 9.

The releases from these facilities appear to be important sources of radionuclides to Lake Ontario. Information concerning these sources should be evaluated further to assess the significance of the releases.

Other facilities include a fuel fabrication facility at Port Hope operated by Zircatec and fabrication facilities in Toronto and Peterborough operated by Canadian General Electric.

The uranium ore concentrates undergo further processing before use in nuclear power plant reactors, including enrichment with ^{235}U before producing the uranium dioxide or metal for fuel elements. Heavy water reactors can use an unenriched uranium concentration; light-water reactors need an enriched uranium (about 2–4%) fuel.

Port Hope is an "Area of Concern" of the RAPs Programs of the Great Lakes Water Quality Agreement. Data from the Port Hope area show that most radionuclide pollutants from fuel fabrication and processing sorb to sediments.

Port Hope is an Area of Concern under the Remedial Action Plan (RAPs) programs of the Agreement. It is the only Area of Concern in which radioactivity is a documented problem causing impaired uses of the resources, and the RAPs documentation contains information on radionuclide levels in various environmental compartments. The Task Force has used the RAP documents as source materials for the Inventory (Krauel et al. 1990).

FIGURE 2. CAMECO PORT HOPE FACILITY ANNUAL AVERAGE URANIUM CONCENTRATIONS — RELEASES OF COOLING WATER FROM NORTH UO_2 AND WEST UF_6 PLANTS

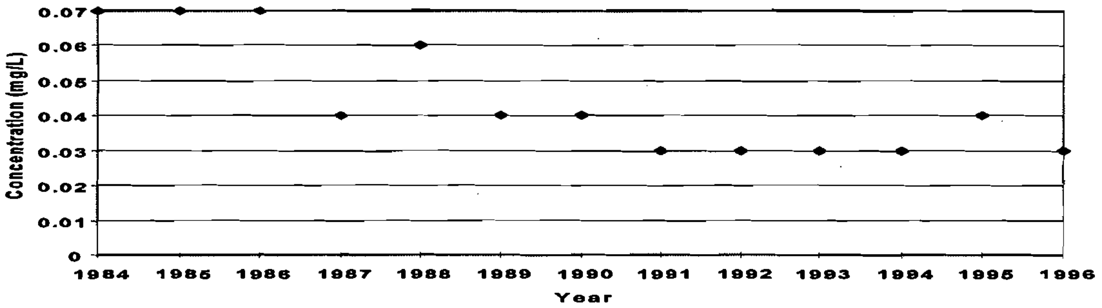


FIGURE 3. CAMECO PORT HOPE FACILITY ANNUAL AVERAGE AIRBORNE URANIUM EMISSIONS

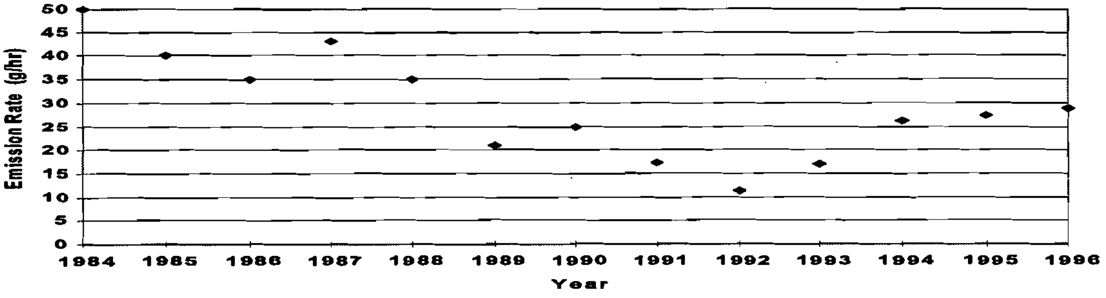


FIGURE 4. CAMECO BLIND RIVER REFINERY ANNUAL AVERAGE URANIUM CONCENTRATIONS — LIQUID RELEASES

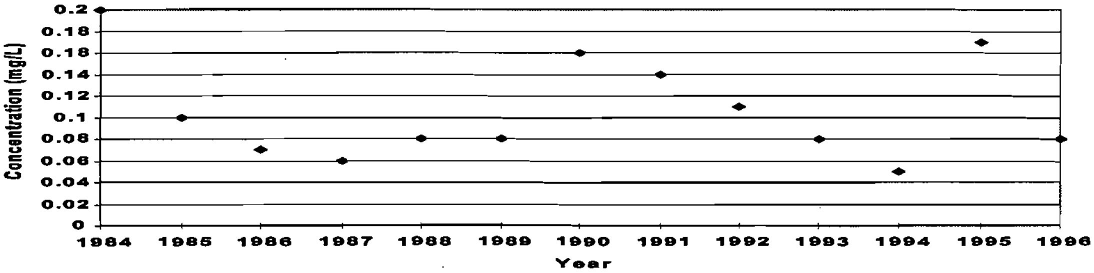


FIGURE 5. CAMECO BLIND RIVER REFINERY ANNUAL AVERAGE AIRBORNE URANIUM EMISSIONS

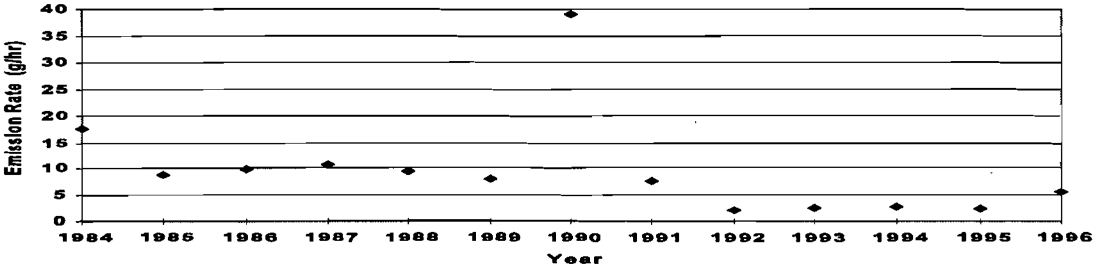


TABLE 8
CAMECO WELCOME LOW-LEVEL WASTE MANAGEMENT FACILITY LIQUID RELEASES

		Radium		Uranium	
Year	Annual Flow (10^6 L)	Avg. Conc. (Bq/L)	Annual Loading (10^6 Bq)	Avg. Conc. (mg/L)	Annual Loading (kg)
1983	71.4	0.04	2.86	—	—
1984	73.9	0.07	5.17	—	—
1985	76.4	0.05	3.82	—	—
1986	91.8	0.06	5.51	—	—
1987	76.3	0.055	4.20	—	—
1988	87.4	0.057	4.98	—	—
1989	78.3	0.057	4.46	0.49	38.4
1990	120.8	0.060	7.25	0.25	30.2
1991	NA	NA	NA	NA	NA
1992	120.2	0.060	7.21	0.33	39.7
1993	151.8	0.058	8.80	0.26	39.5
1994	106.7	0.066	7.04	0.32	34.1
1995	86.6	0.056	4.85	0.28	24.3

Notes: Uranium analyses did not begin until September 1988.
Source of Data: CAMECO annual reports submitted to AECB.
NA, not available.

TABLE 9
CAMECO PORT GRANBY LOW-LEVEL WASTE MANAGEMENT FACILITY LIQUID RELEASES

Year	Annual Flow (10^6 L)	Radium Avg. Conc. (Bq/L)	Radium Annual Loading (10^6 Bq)	Uranium Avg. Conc. (mg/L)	Uranium Annual Loading (kg)
1983	90.2	0.11	9.92	0.63	56.8
1984	81.6	0.12	9.79	0.58	47.3
1985	89.6	0.07	6.27	0.74	66.3
1986	76.0	0.07	5.32	0.74	58.2
1987	83.0	0.07	5.81	0.81	67.2
1988	64.5	0.068	4.39	0.67	43.2
1989	65.5	0.100	6.55	0.70	45.9
1990	71.0	0.100	7.10	0.74	52.5
1991	NA	NA	NA	NA	NA
1992	71.6	0.177	12.67	1.40	100.2
1993	101.3	0.110	11.14	1.30	131.7
1994	72.0	0.128	9.22	1.00	72.0
1995	64.5	0.110	7.10	1.00	64.5

Notes: Source of Data: CAMECO annual reports submitted to AECB.
NA, not available.

The seminal work of Wahlgren et al. (1980) on the transuranics showed that these elements in Lake Michigan strongly bind to sediments. A similar process appears to occur for these elements in Port Hope harbour sediments. Originally the concerns about these sediments focused on elevated levels of heavy metals in the turning basin and west slip. In 1984, Environment Canada and the AECB undertook a joint study, "Benthological, Chemical, Radiological and Chronological Evaluation of Sediments in Port Hope Harbour, Ontario" (McKee et al. 1985). Surficial sediments and sediment cores were studied from various locations for both their heavy metal and actinide contents.

Fuel Reprocessing

Once a reactor has come on line, nuclear fission reactions in the fuel generate power. As the fuel becomes depleted, waste products of fission and activation radionuclides build up in the fuel elements. Some waste products poison the nuclear fission process, requiring eventual removal of a fuel element from service. Because a spent fuel element contains high-level radioactive wastes, it must "cool down" (high-activity radionuclides must decay) on site for periods varying from 6 months to several years before any further processing of the fuel element, such as extraction of unused uranium and plutonium to produce new fuel elements (fuel reprocessing) on-site or off-site, or on-site storage for future disposal or shipment to a high-level waste repository. As of the time of publication of this report, fuel reprocessing is currently not being carried out at facilities in North America.

Fuel reprocessing has several steps, each of which generates high-level and low-level radioactive waste effluents. The steps first separate uranium and plutonium from other radionuclides, and then from each other, and produce gaseous and liquid effluents, which also require further processing before release to the environment. Gaseous effluents contain tritium and radioactive isotopes of iodine, krypton, xenon, ruthenium, and tellurium. Liquid effluents usually contain isotopes of rare earth elements, cesium, and a few actinides. Special processes aim to recover and remove the iodine radionuclides, especially ^{129}I (half-life of 16 million years). The liquid wastes are usually concentrated through evaporation, and stored in underground tanks for eventual disposal as high-level waste.

Although the radionuclides expected in reprocessing effluents are known, the Task Force lacked sufficient quantitative data for an inventory of the radionuclides. However, fuel reprocessing is not considered a major source of radioactivity relative to the nuclear power plants. As the Task Force receives additional data on reprocessing effluents, it will consider supplements to the inventory report.

A facility once used for fuel reprocessing plant at West Valley, New York, now serves as a low-level waste repository.

The Western New York Nuclear Service Center (West Valley) once operated as a fuel reprocessing plant. It was the only reprocessing plant in the Basin. The facility ceased the reprocessing in 1972, but continued as a storage site for its own locally produced high-level and low-level wastes, and to receive low-level wastes from other facilities. In 1982, the United States Department of Energy (DOE) proposed a long-term management strategy for the nuclear wastes on the site. DOE proposed to concentrate, chemically treat, and convert the liquid high-level wastes to a solid form suitable for transportation off site and permanent placement in a federal geological repository. In 1988, DOE proposed a project with "alternatives" to complete closure or long-term management of the facilities and, in 1995, proposed an implementation plan for the various alternatives. In 1996, DOE announced availability of a Draft Environmental Impact Statement (61 Federal Register 11620) for "Completion of the West Valley Demonstration Project and Closure or Long-Term Management of Facilities at the Western New York Nuclear Service Center."

The New York State Energy Research and Development Agency (NYSERDA) owns the West Valley site on behalf of the "taxpayers of New York." The site receives low-level radioactive wastes and has some wastes from industrial activities, and generates other wastes. The proposed decommissioning alternatives affect the estimation of radionuclide inventories and may signal current and future thinking by government agencies on decommissioning strategies, especially in a period of very tight and highly constrained budgets. The proposed decommissioning alternatives for West Valley appear in Table 10. As of the time of this report, the decision from DOE was pending. The Environmental Impact Statement does not indicate a DOE or NYSERDA preferred alternative.

TABLE 10
PROPOSED ALTERNATIVES FOR THE DECOMMISSIONING OF
WESTERN NEW YORK NUCLEAR SERVICE AT WEST VALLEY, NEW YORK
(adapted from 61 Federal Register 11620 - March 21, 1996)

1 **Removal and Release to Allow Unrestricted Use.** This alternative provides for complete removal of all facilities, buried wastes, and residuals, to assure "minimal" remnants of nuclear operations. It then permits an "unrestricted" civilian use of the site by the owners, New York State Energy Research and Development Authority (NYSERDA).

2 **Removal, On-Premises Waste Storage, and Partial Release to Allow Unrestricted Use.** This alternative provides for the removal of existing facilities including buried waste, but does not remove on-site stored high-level, low-level and low-level mixed (with hazardous materials) waste. Hazardous and industrial (non-radioactive) waste would be disposed of off-site.

3 **In-Place Stabilization and On-Premises Low-Level Waste Disposal.** This alternative provides the in-place stabilization of contaminated structures and buried wastes. Uncontaminated structures are removed. Low-level waste would continue to be disposed of on-site, and all other waste would be disposed of off-site.

4 **Monitoring and Maintenance Only.** This is a "no action type of alternative," which provides for the management of the site in its current configuration and condition. Only hazardous wastes would be disposed of off-site.

5 **Discontinue Operations.** This alternative stops any current activities on the site, and leaves the site in its current configuration and condition. All wastes currently on-site remain on-site. No site closure activities and no provision for monitoring.

Notes: (1) Alternative 3 continues the present use of the facility as a low-level nuclear waste repository.
(2) Alternative 4 meets a minimum regulatory requirement under the National Environmental Protection Act (NEPA) and assures a monitoring baseline for comparing the environmental effects from "action alternatives."
(3) Alternative 5, "Do Nothing," provides a worst-case baseline for comparison with other alternatives. Public comments on previous proposals of this alternative have led DOE and NYSERDA **not** to consider this alternative as "reasonable," but neither DOE nor NYSERDA have officially rejected it.

2.3.3 Emissions from Nuclear Power Plants in the Great Lakes Basin

Nuclear power in the Great Lakes Basin dates from 1962 and the commissioning of the Big Rock Point Plant. In 1997, there are 15 facilities. Decommissioning of reactors may begin as early as 2000 with the expiration of the license for Big Rock Point. Table 11 presents data on the licensed nuclear power plants in the Basin. As of the time of this report, Ontario Hydro has taken seven of its reactors off-stream and may consider decommissioning them. This unexpected action occurred in late August 1997.

There are three kinds of nuclear power plants in the Basin: two kinds of light-water reactors (LWR) and heavy-water reactors (HWR). The LWR systems are the "pressurized water reactor" (PWR) and "boiling water reactor" (BWR). The United States facilities are all LWR systems and the Canada facilities are all HWR systems. The systems' names describe the reactor cooling and moderating systems used. A fourth type of reactor found in North America at university and hospital research laboratories is the gas-cooled reactors (GCR) but is not used for electric power production. Under development is a fast-breeder reactor (FBR). The nuclear power plant, Fermi 1, had a FBR system, but the reactor was decommissioned following an accident and replaced by Fermi 2, a reactor of the BWR type.

TABLE 11
NUCLEAR POWER PLANT REACTORS IN THE GREAT LAKES BASIN

Reactor	Start-Up (Year)	Net Electrical Power (MWe)	Reactor Type	License Expiration (Year)
		United States		
Big Rock Point	1962	70	BWR	2000
Nine Mile Point 1	1969	625	BWR	2009
R.E. Ginna	1970	420	PWR	2009
Point Beach 1	1970	497	PWR	2010
Palisades	1971	700	PWR	2007
Point Beach 2	1972	497	PWR	2013
Zion 1,2	1973	2 × 1050	PWR	2013
D.C. Cook 1	1975	1050	PWR	2014
Kewaunee	1974	520	PWR	2013
J.A. Fitzpatrick	1975	800	BWR	2014
Davis–Besse 1	1977	910	PWR	2017
D.C. Cook 2	1978	1050	PWR	2017
Fermi 2	1985	1090	BWR	2025
Perry	1986	1205	BWR	2026
Nine Mile Point 2	1987	1070	BWR	2026
		Canada		
Douglas Point	1966	220	HWR	Shut Down
Pickering A	1971/1973	4 × 508	HWR	1998
Pickering B	1983/1984	4 × 508	HWR	1998
Bruce A	1976/1979	4 × 750	HWR	1998
Bruce B	1984/1987	4 × 840	HWR	1999
Darlington A	1990/1993	4 × 850	HWR	1998

Notes: (1) Sources of data: U.S. NRC Information Digest, 1995 Edition; Ahier and Tracy (1995), "Radionuclides in the Great Lakes Basin," published; UNSCEAR (1977, 1982, 1988); Reporter, AECB Newsletter, Spring 1996.
(2) BWR: boiling water reactor; PWR: pressurized water reactor; HWR: heavy water reactor
(3) The Douglas Point reactor is no longer operating but has not yet been decommissioned.

The relative quantities radionuclides produced depend on the reactor type, including the technology and materials of construction, the amount of electricity generated, and the processes used to handle effluents and waste products. UNSCEAR documents present nuclear power plant data in a format based on reactor type and power production. UNSCEAR also applies a special averaging technique, normalization, which puts radionuclide production in a reactor on a unit energy basis, averaged over all reactors worldwide of the given type. The Task Force calculated normalization of the data but did not find it useful.

Data on atmospheric emissions from the nuclear power plants in the Great Lakes show varying degrees of completeness, specificity, and descriptive information. All power plants report particulate matter, tritium, total β-emissions excluding tritium, and ^{131}I. Some plants report "total noble gases," α and γ radiation; other plants also list specific noble gas radionuclides (*e.g.*, ^{41}Ar, ^{85}Kr, ^{133}Xe, ^{135}Xe); and other plants report other radionuclides of iodine (*e.g.*, ^{133}I, ^{134}I, and ^{135}I). The measurement of xenon radionuclides depends on their energy spectrum: those emitting γ radiation below 1 MeV might not be reported. Canadian (thus HWR) plants generally report fewer radionuclides, but Canadian regulations consider the effect of all radionuclides migrating to humans through all pathways. The Task Force learned that while many radionuclides do not require reporting, sometimes the power plant authorities collect this information, but without any consistency or regularity.

The following radionuclides receive consistent reporting in the atmospheric and aquatic emissions from most United States and Canadian facilities: tritium (^{3}H), strontium (^{90}Sr), iodine (^{131}I), cesium (^{134}Cs, ^{137}Cs), and noble gases (a mixture of radioactive isotopes of krypton and xenon). Table 12 summarizes the cumulative amount of each radionuclide released from the three reactor types by the air and water pathways. The results have been summed over all lakes. Tritium and noble gases dominate the radioactivity of atmospheric releases; tritium also dominates the radionuclides in aquatic releases. The emission of the six major radionuclide types are discussed below.

Tritium

The six graphs that compose Figure 6 show the emissions over time for tritium from the three reactors types and for the air and water pathways. Summations have been carried out over all lakes. All results are expressed in terabecquerels (1 TBq = 10^{12} Bq or 27 Ci). When comparing graphs, note that the scales vary.

The information shows that, throughout the entire time period, tritium emissions from HWRs consistently exceed by two orders of magnitude those from PWRs and exceed by three orders of magnitude those from BWRs. Further, the HWRs show a clearly increasing trend with time in both the airborne and waterborne emissions. This comports with expectations for HWRs. Deuterium in the heavy water readily captures neutrons from the fission process to become tritium. As the heavy water "ages," tritium levels increase in the water. If operating conditions do not change, the emissions of tritium will gradually increase in proportion to their concentration in the water. However, the high emissions for these reactors, such as for airborne tritium in 1981, 1983, and 1989 and for waterborne tritium in 1987, 1988, 1989, 1991, and 1992, do not reflect a gradual increase or an increase in the number of reactors operating but rather anomalous releases. The Task Force has learned that tritium is currently removed from all Ontario HWRs at the Darlington facility. This facility also releases tritium to the Great Lakes airshed. Also, HWR generating capacity increased considerably over the time period: 140% compared with an increase of only 30% for PWRs and BWRs. Time trends are much less apparent in the emissions from the PWRs and BWRs. Occasional elevated values from these reactor types occur in the earlier years. Explanations for these anomalies require review of the detailed reactor records.

The two graphs that compose Figure 7 show the cumulative emissions of tritium for both the airborne and waterborne pathways. The units are petabecquerels (1 PBq = 10^{15} Bq or 27,000 Ci). These results were obtained by integration of the year-by-year data from the previous graphs, then summing over the three reactor types, and correcting each radionuclide for subsequent disappearance through radioactive decay. While this simplistic approach ignores the disappearance of tritium from the Great Lakes ecosystem through dispersion, drainage, or incorporation into sediments, the graphs do show the total burden of tritium placed upon the global biosphere attributable to reactor emissions in the Basin.

Quite simply, the tritium must go somewhere until its final transformation into stable helium-3 by radioactive decay. Table 13 shows tritium production and emission for different types of nuclear power plant technologies.

Strontium-90

The four graphs that compose Figure 8 show the ^{90}Sr emissions. The Task Force obtained data only for the United States reactors (PWRs and BWRs). The units here are megabecquerels (1 MBq = 10^6 Bq or 27 microcuries (µCi)). No clear time trends appear in the emission data. Some early data on BWRs in the 1970s showed a few anomalously high values. The PWRs showed an elevated value in 1984 for the airborne emissions, and in 1990 for the waterborne emissions.

The two graphs that compose Figure 9 show cumulative emissions of ^{90}Sr analogously to Figure 7. With a half-life of about 30 years, the ^{90}Sr will slowly decay from the global biosphere, provided the absence of new ^{90}Sr releases.

TABLE 12
CUMULATIVE AIRBORNE AND WATERBORNE EMISSIONS OF THE MAJOR RADIONUCLIDES FROM THE THREE REACTOR TYPES (1980–1993)

	BWR	PWR	HWR	Total
Airborne (GBq)				
^{3}H	54,289	355,750	57,770,000	58,180,030
^{90}Sr	0.21568	0.05305	*	0.26873
^{131}I	274.01	64.62	29.32	367.95
^{134}Cs	5.79	6.95	*	12.74
^{137}Cs	3.14	8.86	*	12
Noble gas			*	1.463×10^9
Waterborne (GBq)				
^{3}H	11,840	2,316,160	41,010,000	43,338,000
^{90}Sr	3.09	7.29	*	10.38
^{131}I	36	209.7	*	245.7
^{134}Cs	245.96	241.58	*	487.54
^{137}Cs	507.04	348.72	*	855.76

*These radionuclides are emitted but not separately reported in Canada.

TABLE 13
TRITIUM PRODUCTION AND EMISSIONS FOR DIFFERENT TYPES OF NUCLEAR POWER PLANT TECHNOLOGIES

	Tritium Production Rate in Reactors by		Tritium Emission Rate from Reactors	
Reactor Type	**Fission**	**Neutron activation**	**to air**	**to water**
PWR				
Ci·(GWe·year)$^{-1}$	$(1.4–2.0) \times 10^4$	$(0.1–8.0) \times 10^2$	1.0×10^2	9.0×10^2
TBq·(GWe·year)$^{-1}$	7.5×10^2	40	4	33
BWR				
Ci·(GWe·year)$^{-1}$	$(1.4–2.0) \times 10^4$	Negligible	60	1.4×10^2
TBq·(GWe·year)$^{-1}$	7.5×10^2	Negligible	2	5
HWR				
Ci·(GWe·year)$^{-1}$	$(1.4–2.0) \times 10^4$	$(0.6–2.4) \times 10^6$	$(0.3–1.7) \times 10^4$	$(2.0–7.0) \times 10^3$
TBq·(GWe·year)$^{-1}$	7.5×10^2	9.0×10^4	6.0×10^2	1.5×10^2

Notes: (1) Fission production of tritium does not depend on the technology in the PWR, BWR, and HWR reactors. The three reactor types should produce equal levels of tritium by fission on per unit energy basis.

(2) Units are either curies or terabecquerels per gigawatt-year (the number of gigawatts of power output multiplied by the number of years to standardize the values across reactor types).

(3) Tritium production in the boron-based control rods in a PWR is reflected in the lower limit of the range under neutron reaction; tritium production in the boron-based control rods of the BWR is negligible and is not included. Tritium production by neutron reaction in the HWR occurs via the tritium–deuterium reaction of heavy water.

(4) All data in the table are based on actual measurements at power plants. The original source indicates data which were inferred or estimated by other means.

(5) Source: IAEA (1980).

Iodine-131

The five graphs that compose Figure 10 show the reported emissions for ^{131}I. The data show no clear trends, but rather some anomalous emissions in the earlier years. Airborne emissions of ^{131}I are highest for the BWRs, although all three reactor types show some releases. In the BWR, the primary coolant boils to produce steam for the turbines. A volatile radionuclide, such as ^{131}I, is readily released from the coolant to the atmosphere. Waterborne releases of ^{131}I are much less. The Task Force did not generate a cumulative graph for this radionuclide, since its half-life of only 8 days assures that it does not accumulate in the biosphere.

Cesium-134 and Cesium-137

Cesium emission data are not available from the HWRs. Figures 11 and 12, which are each composed of four graphs, show the emissions of both isotopes of cesium for the BWRs and PWRs, respectively. The display of the results reflects the fact that often the emissions of both isotopes occur in parallel. The units are in gigabecquerels (1 GBq = 10^9 Bq, or 27 millicuries (mCi)). Cesium is somewhat more volatile than strontium; thus, its emissions are higher than strontium. Except for a few anomalies in the early years, the data do not show any time trends in the cesium emissions.

^{134}Cs (half-life: 2.05 years) is removed fairly rapidly from the biosphere. The two graphs that compose Figure 13 show the cumulative emissions for the longer lived ^{137}Cs (half-life: 30 years). The slow decrease in the ^{137}Cs burden with time means that the emissions have been fairly steady, and that the isotope disappears rather slowly from the biosphere.

Noble Gases

Figure 14 shows the noble gas emissions to air for the combined BWRs and PWRs. Generally, emissions were higher before 1984 than after that time, with one anomalously high value in 1975. Since noble gases are not water soluble, their emissions to water are not considered significant. Most radionuclides of noble gases decay by β-particle emission to elements that form oxides and hydroxides, either in particulate or aerosol form. Other non-gaseous fission and activation radionuclides can form aerosols, which accompany airborne effluents. The aerosols and particulates become part of atmospheric fallout. Monitoring data are available both for "total noble gases" and total particulates in gaseous emissions. Air pollution control systems at nuclear power plants prevent the emissions of all but the very finest particulates.

Short-lived xenon isotopes account for most of the emissions , assuring essentially no build-up. The one exception is ^{85}Kr (half-life: 10 years) which remains in the atmosphere and becomes globally dispersed.

Other Radionuclides, Including ^{14}C and ^{129}I

^{14}C (half-life: 5730 years) and ^{129}I (half-life: 16 million years) are also important because of their long half-lives, but are not routinely reported by most facilities. ^{129}I is difficult to measure; however, a new technique involving accelerator mass spectometry may make this possible in the future. This technology is now available at the University of Toronto. Tables 14 and 15 give information on carbon-14 production and emissions.

TABLE 14
CARBON-14 PRODUCTION AND EMISSIONS FOR DIFFERENT TYPES OF NUCLEAR POWER PLANT TECHNOLOGIES

	^{14}C Production Rate			^{14}C Emission Rate
Reactor Type	Fuel	Moderator and Coolant	Total	From the Reactor to Air
PWR				
Ci·(GWe·year)$^{-1}$	4–12	1–11	6–50	5–13
kBq·(GWe·year)$^{-1}$	0.4	0.3	0.7	0. 3
BWR				
Ci·(GWe·year)$^{-1}$	11–16	4–11	15–50	11–26
kBq·(GWe·year)$^{-1}$	0.6	0.4	1.0	0.4
HWR				
Ci·(GWe·year)$^{-1}$	20–50	320–550	450–560	270
kBq·(GWe·year)$^{-1}$	1.0	20	21	10

Notes: (1) Totals reflect direct measurements from several sources not all of which quantified both fuel and moderator and coolant.

(2) Units are either curies or kilobecquerels per gigawatt-year (the number of gigawatts of power output produced multiplied by the number of years to standardize the values across reactor types).

(3) Most of the ^{14}C is emitted as radioactive carbon dioxide or radioactive methane gas. Thus, only gaseous emissions to the atmosphere appear in the table. The incorporation of radioactive carbon into molecules in the liquid effluents is less than 1% according to the original source material.

(4) All data are based on actual measurements at power plants.

(5) Source: IAEA (1980).

TABLE 15
PRODUCTION OF ^{14}C IN NUCLEAR REACTORS
(adapted from Tait et al. 1980)

Fission production: 1.84×10^{-5} moles ^{14}C per kilogram of uranium initially in the fuel or 1.11×10^{19} atoms/kg uranium.

Total carbon isotope production (^{12}C, ^{13}C and ^{14}C) in the fuel: 8.47×10^{-3} moles of carbon per kilogram of uranium initially in the fuel or 5.1×10^{21} atoms/kg uranium.

Relative ^{14}C production: <0.2% of the carbon isotopes.

Activity of ^{14}C produced: 4.26×10^{7} Bq per kilogram of initial uranium.

Production of ^{14}C relative to other radionuclides produced from impurities: 11 of 64

Fraction of activity of ^{14}C relative to all radionuclides produced from fuel impurities: 0.000968%

Reported Airborne Tritium Emissions for All Lakes (in TBq)

Reactor Type: BWR

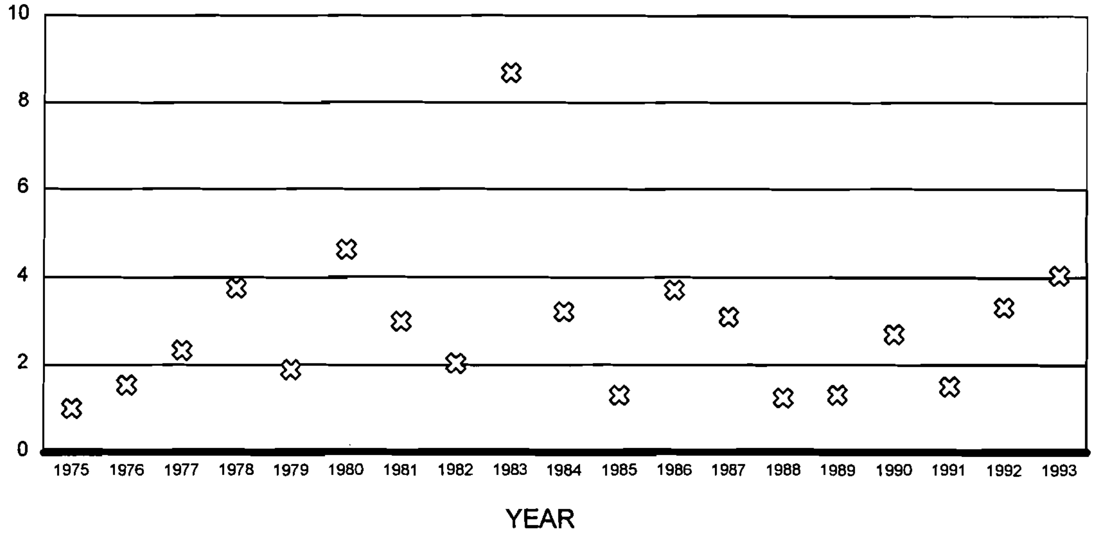


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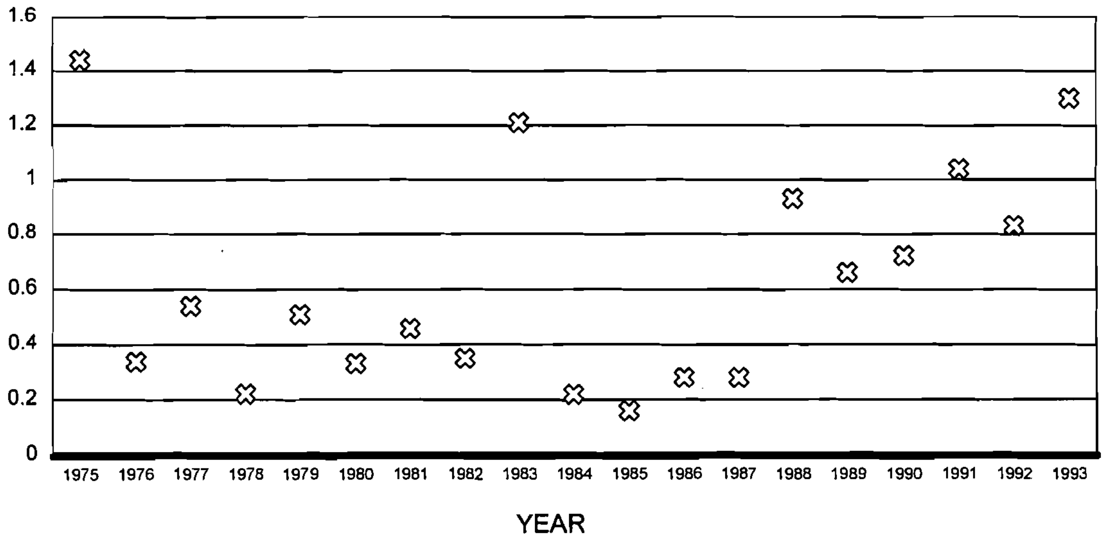
Figure 6

Reported Waterborne Tritium Emissions for All Lakes (in TBq)

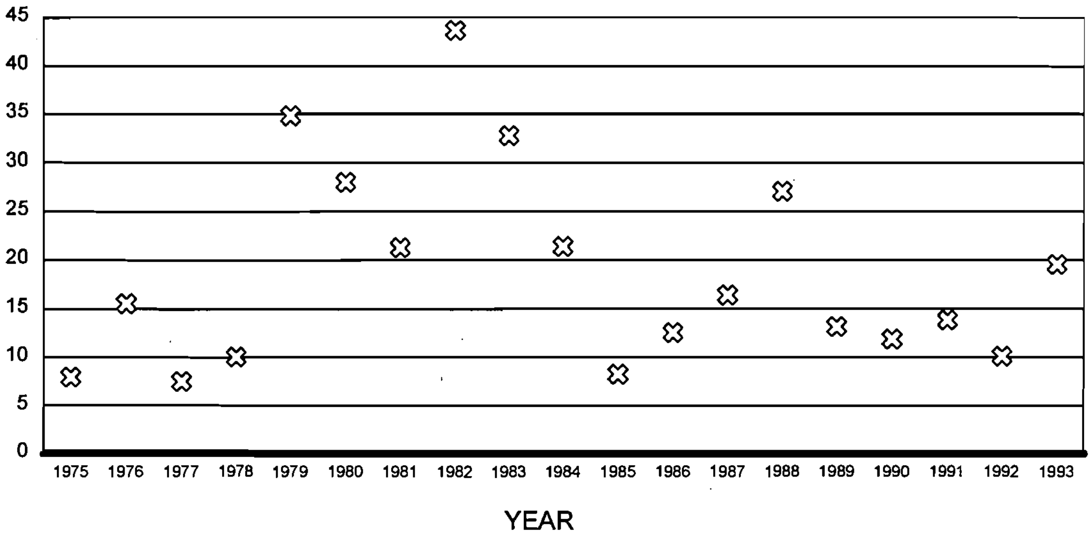
Reactor Type: BWR



Date run: 12/22/97 GRAPH_1

Reported Airborne Tritium Emissions for All Lakes (in TBq)

Reactor Type: PWR

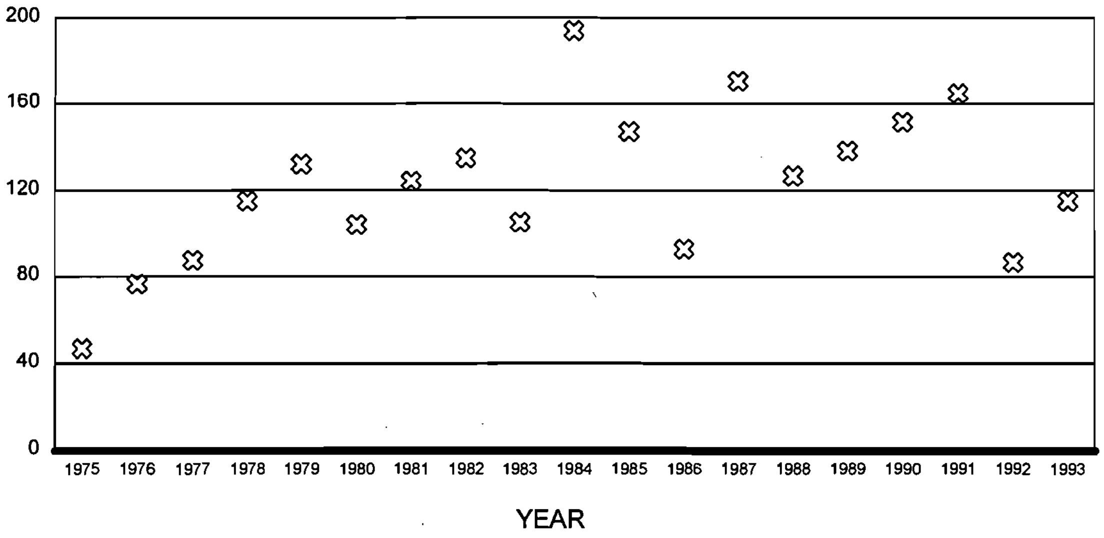


Date run: 12/22/97 GRAPH_1

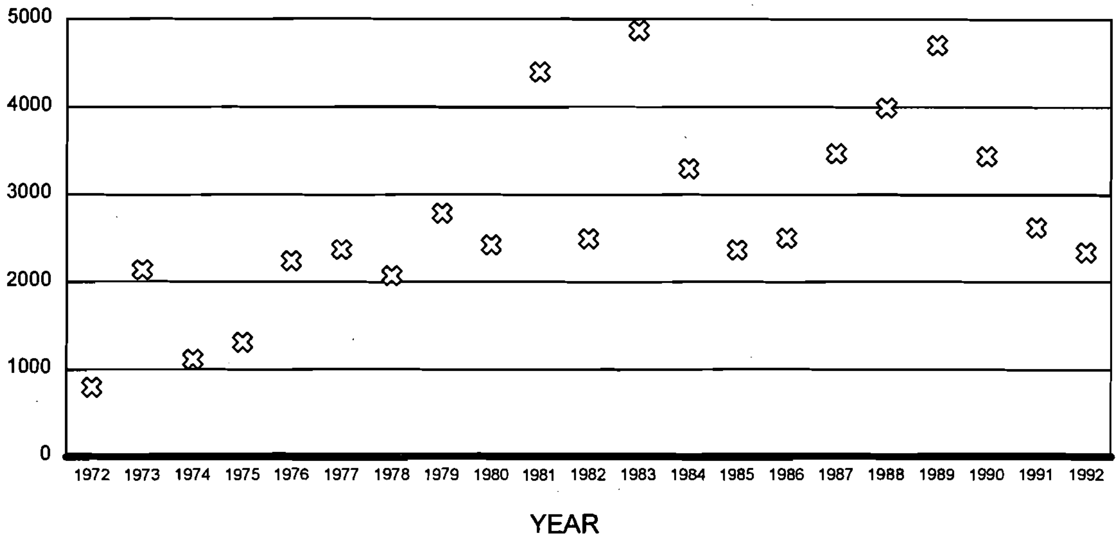
Figure 6 cont.

Reported Waterborne Tritium Emissions for All Lakes (in TBq)

Reactor Type: PWR



Date run: 12/22/97 GRAPH_1

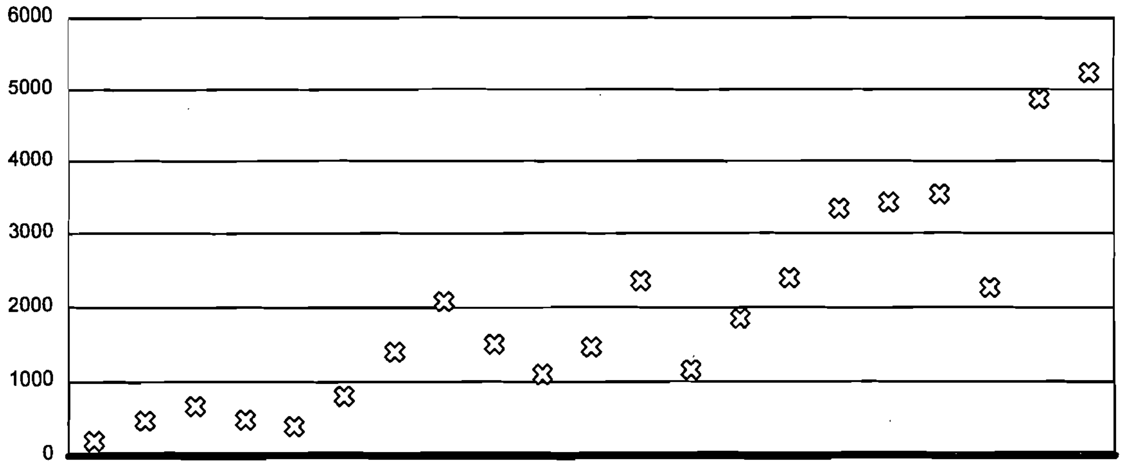


Reported Airborne Tritium Emissions for All Lakes (in TBq)

Reactor Type: HWR

Date run: 12/22/97 GRAPH_1

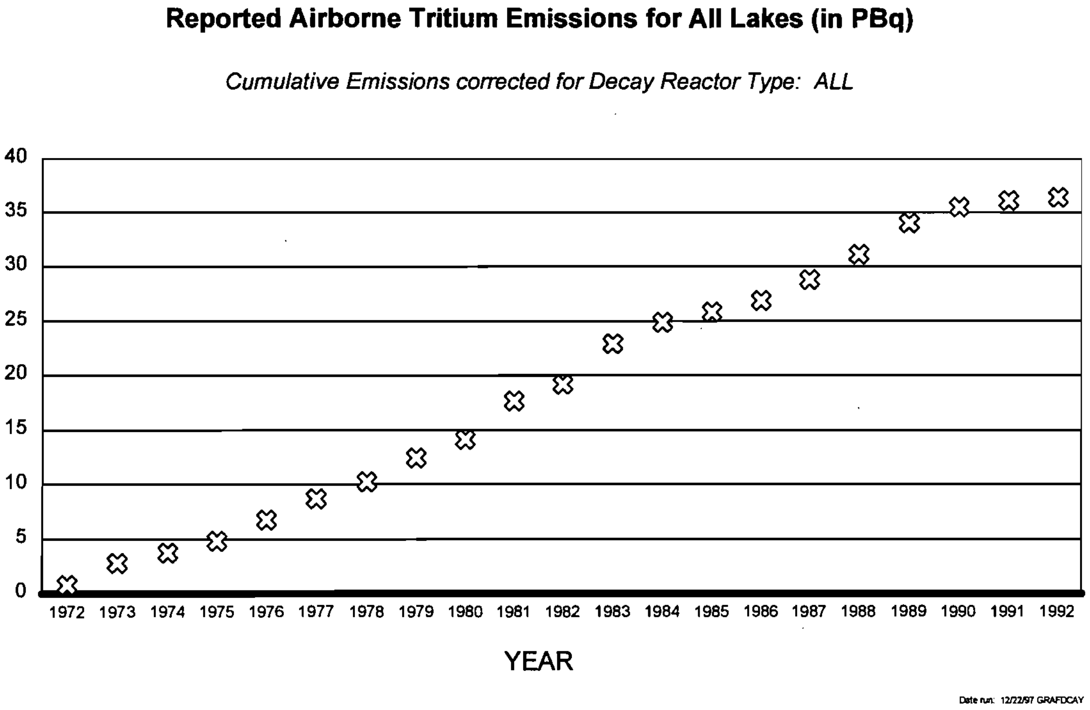
Figure 6 cont.



Reported Waterborne Tritium Emissions for All Lakes (in TBq)

Reactor Type: HWR

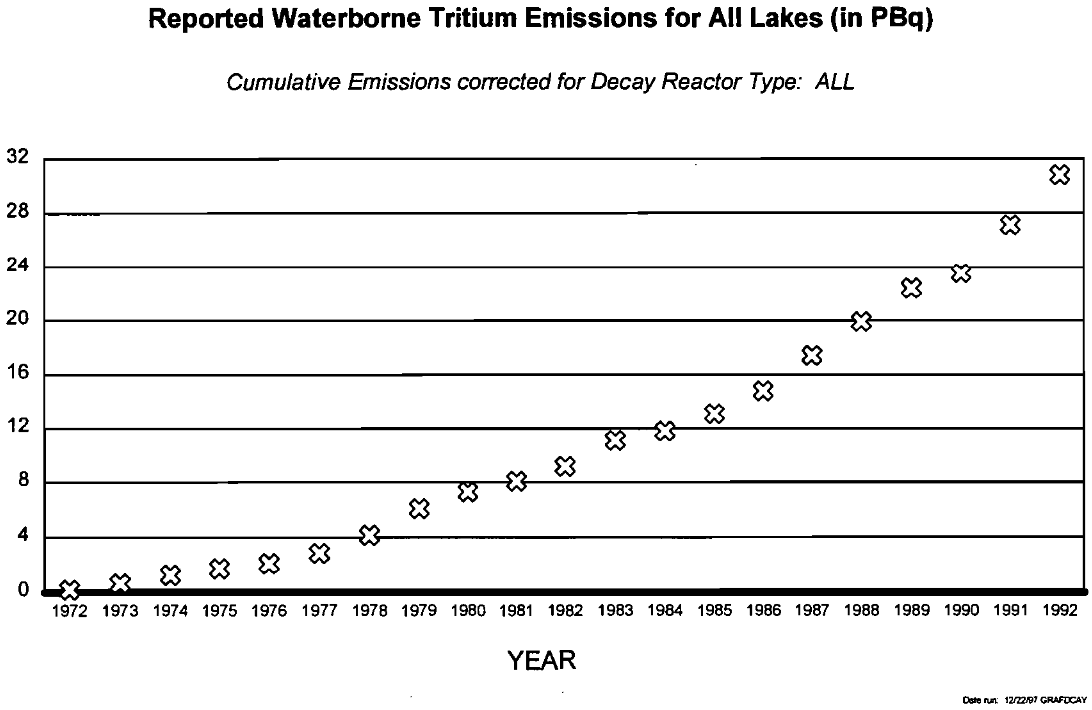
Date run: 12/22/97 GRAPH_1



Reported Airborne Tritium Emissions for All Lakes (in PBq)

Cumulative Emissions corrected for Decay Reactor Type: ALL

Figure 7



Reported Waterborne Tritium Emissions for All Lakes (in PBq)

Cumulative Emissions corrected for Decay Reactor Type: ALL

Reported Airborne Sr-90 Emissions for All Lakes (in MBq)

Reactor Type: BWR

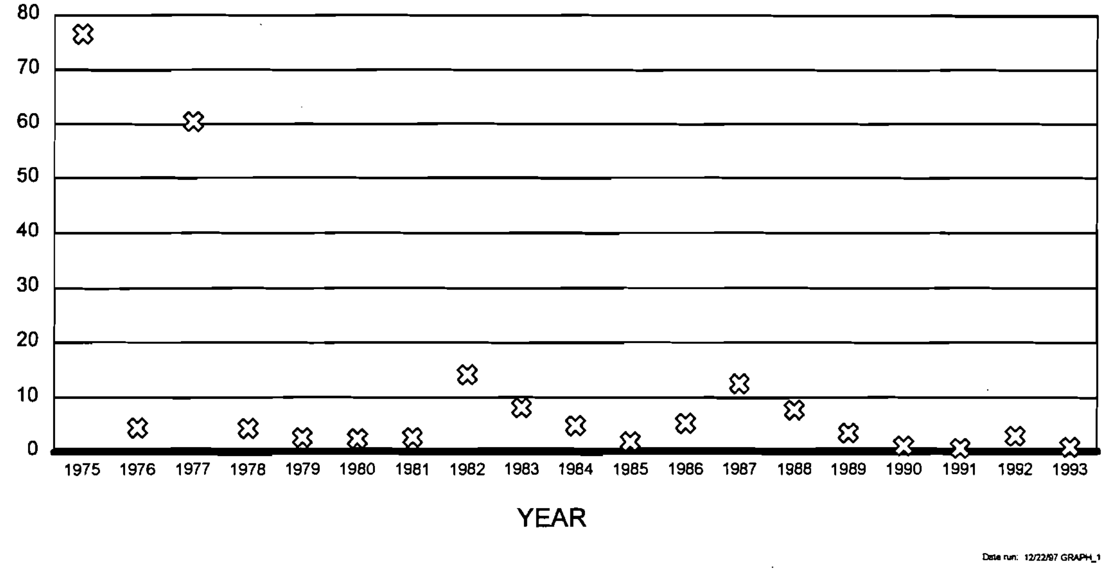
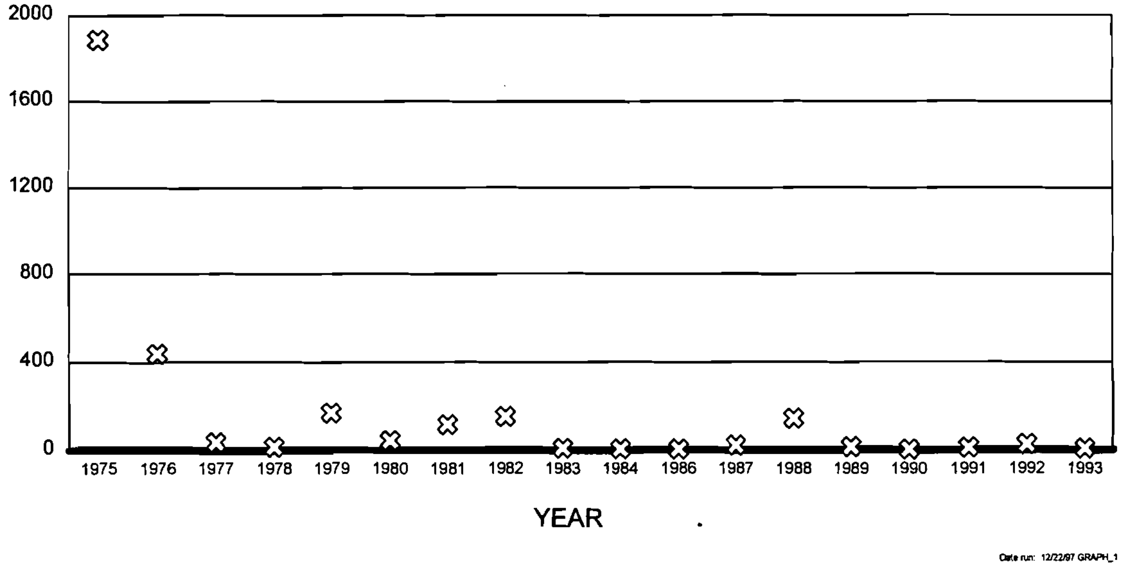
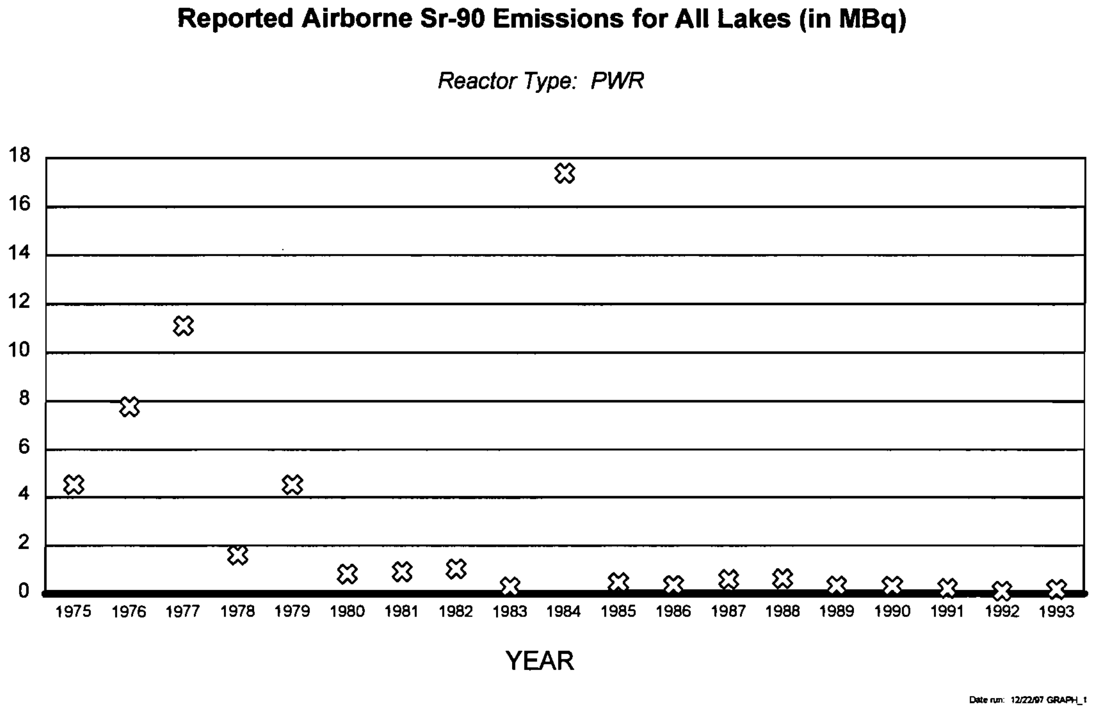


Figure 8

Reported Waterborne Sr-90 Emissions for All Lakes (in MBq)

Reactor Type: BWR

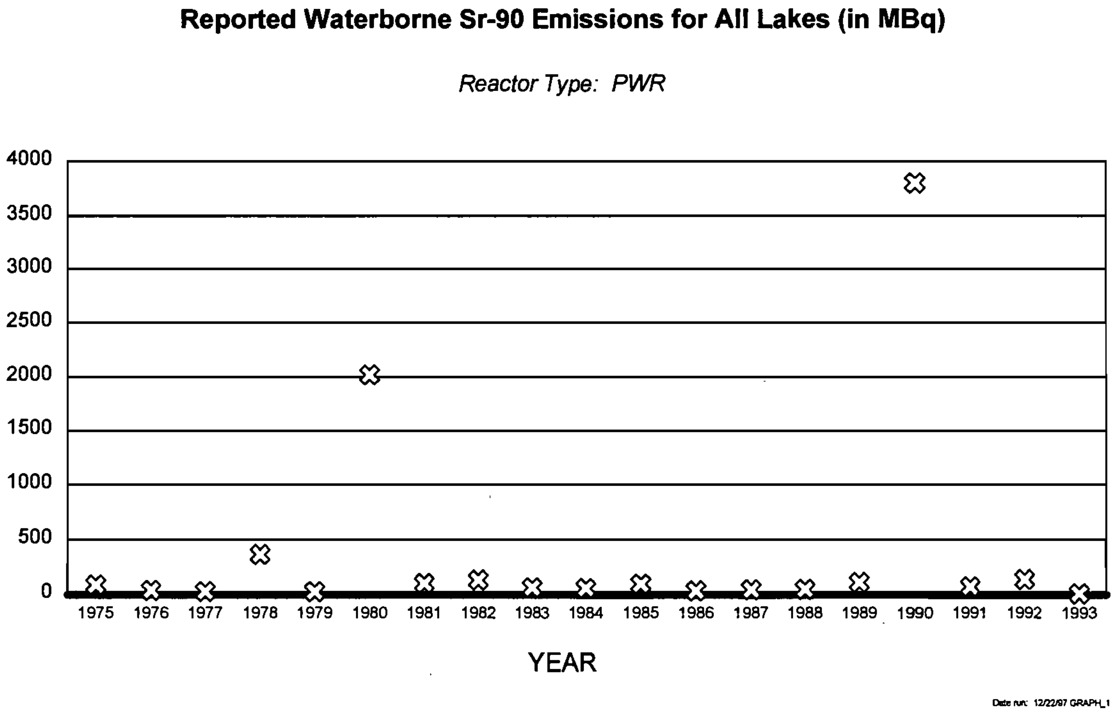




Reported Airborne Sr-90 Emissions for All Lakes (in MBq)

Reactor Type: PWR

Figure 8 cont.



Reported Waterborne Sr-90 Emissions for All Lakes (in MBq)

Reactor Type: PWR

Reported Airborne Sr-90 Emissions for All Lakes (in MBq)

Cumulative Emissions corrected for Decay Reactor Type: ALL

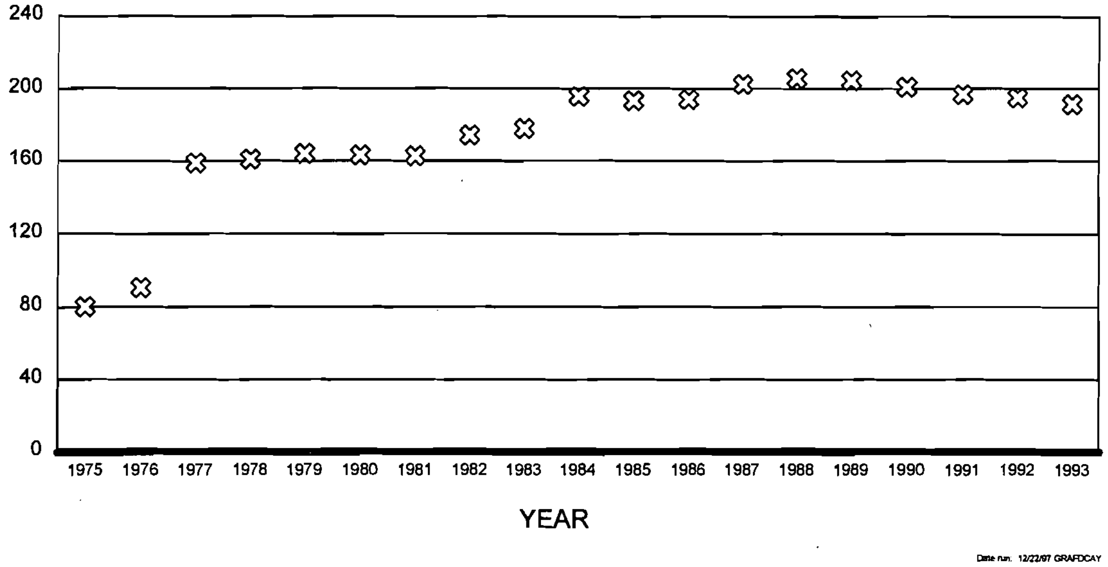
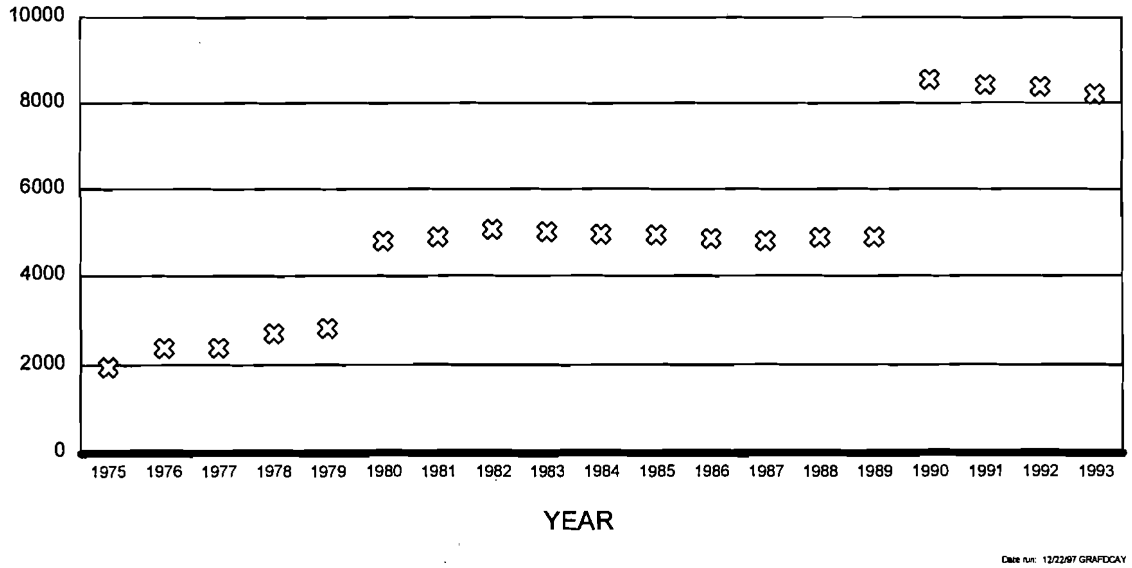


Figure 9

Reported Waterborne Sr-90 Emissions for All Lakes (in MBq)

Cumulative Emissions corrected for Decay Reactor Type: ALL



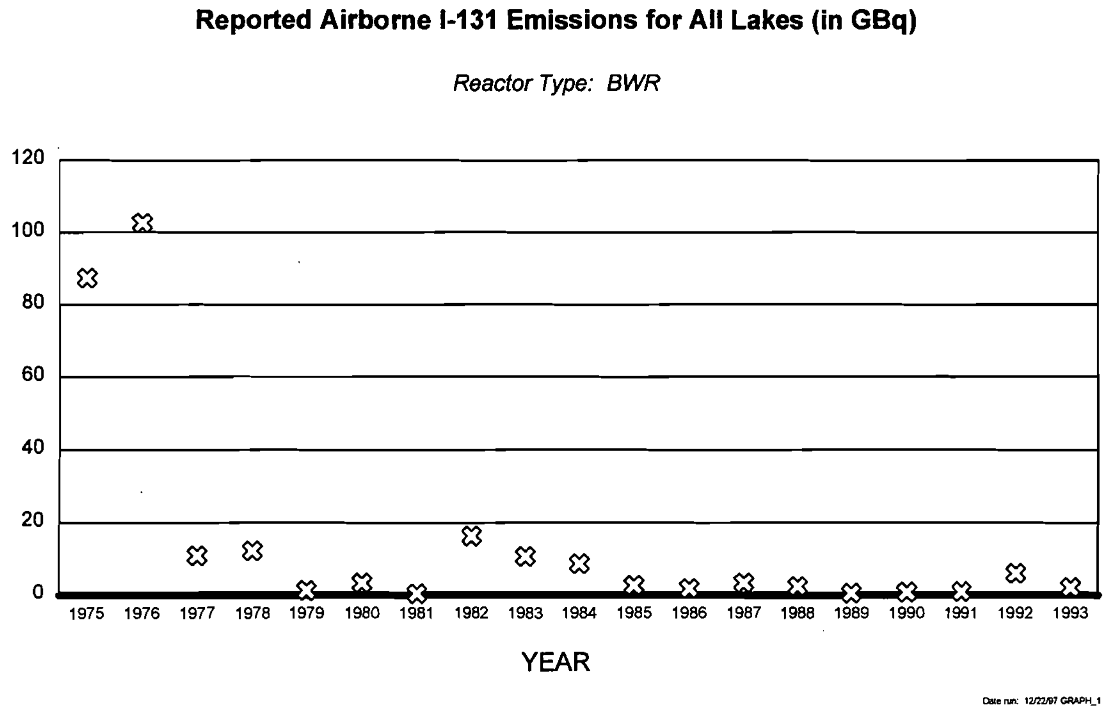
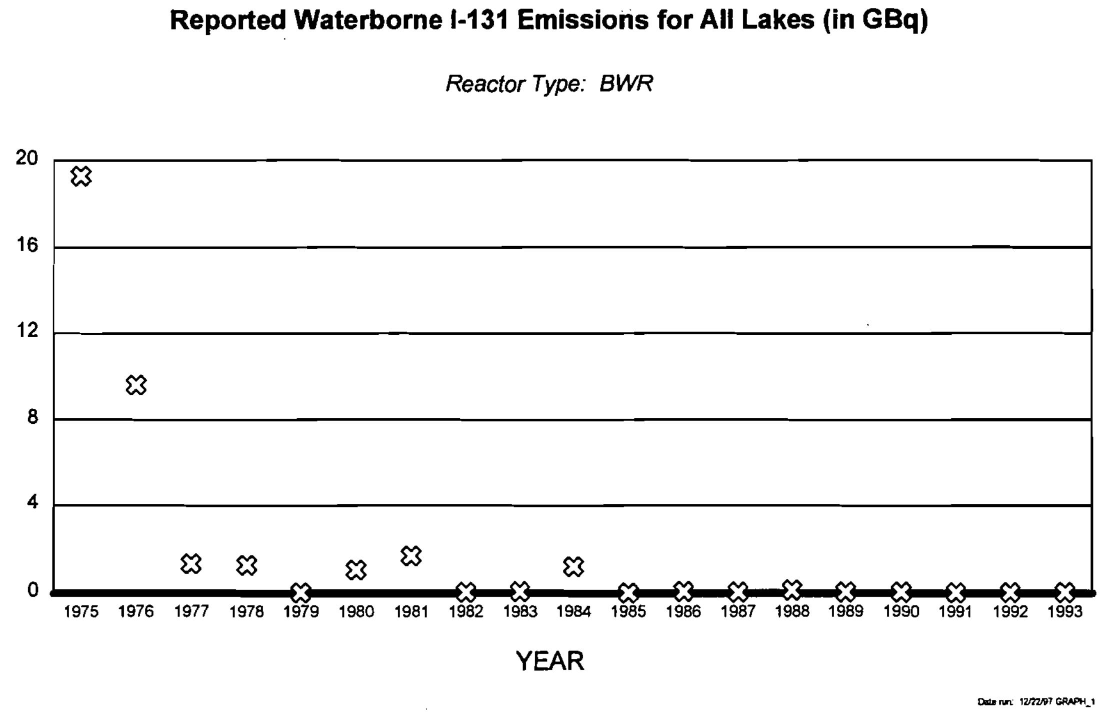
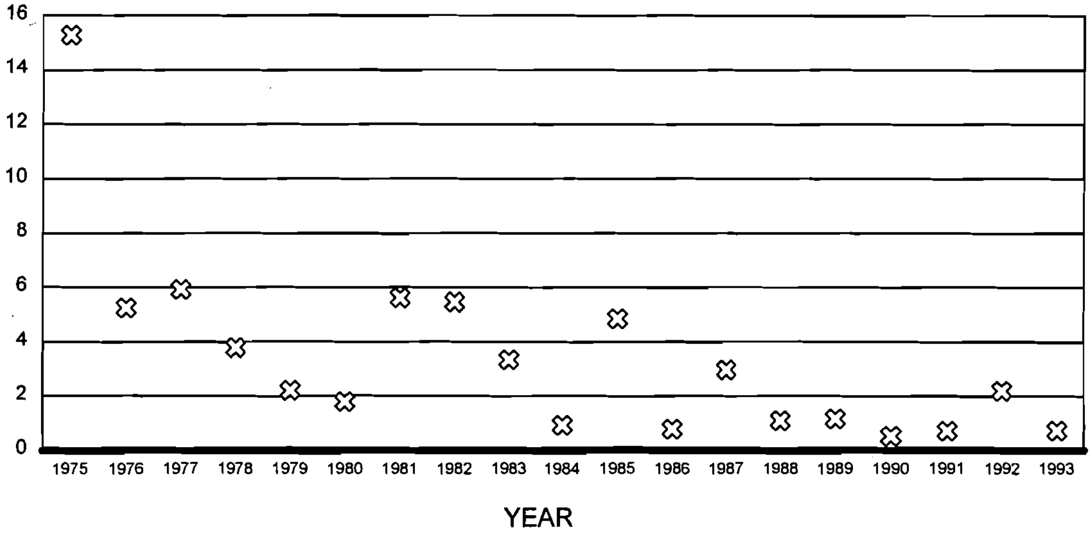

Reported Airborne I-131 Emissions for All Lakes (in GBq)
Reactor Type: BWR
120
100
80
60
40
20
0
1975
1976
1977
1978
1979
1980
1981
1982
1983
1984
1985
1986
1987
1988
1989
1990
1991
1992
1993
YEAR
Date run: 12/22/97 GRAPH_1

Figure 10


Reported Waterborne I-131 Emissions for All Lakes (in GBq)
Reactor Type: BWR
20
16
12
8
4
0
1975
1976
1977
1978
1979
1980
1981
1982
1983
1984
1985
1986
1987
1988
1989
1990
1991
1992
1993
YEAR
Date run: 12/22/97 GRAPH_1

Reported Airborne I-131 Emissions for All Lakes (in GBq)

Reactor Type: PWR

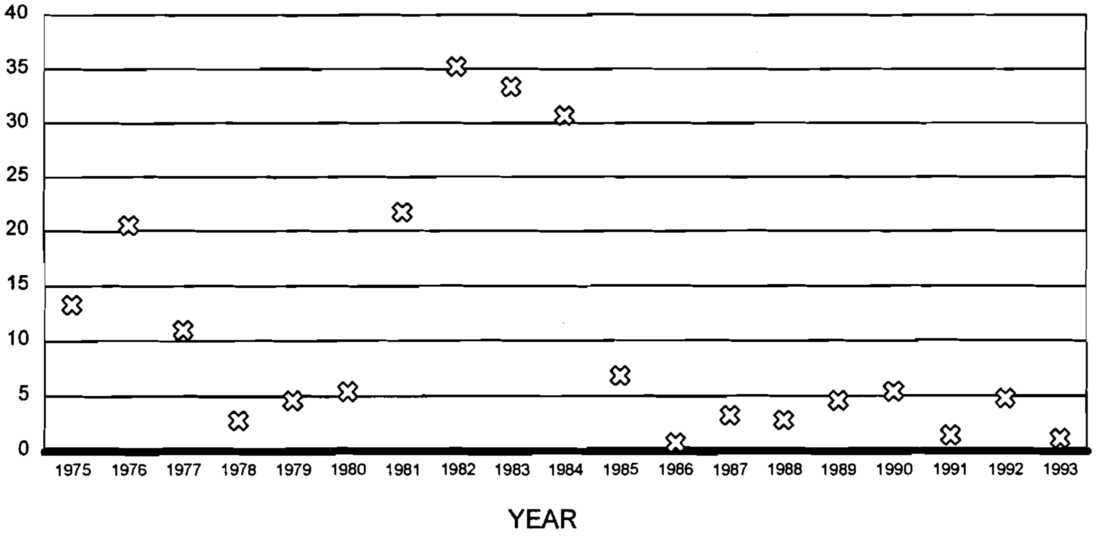


Date run: 12/22/97 GRAPH_1

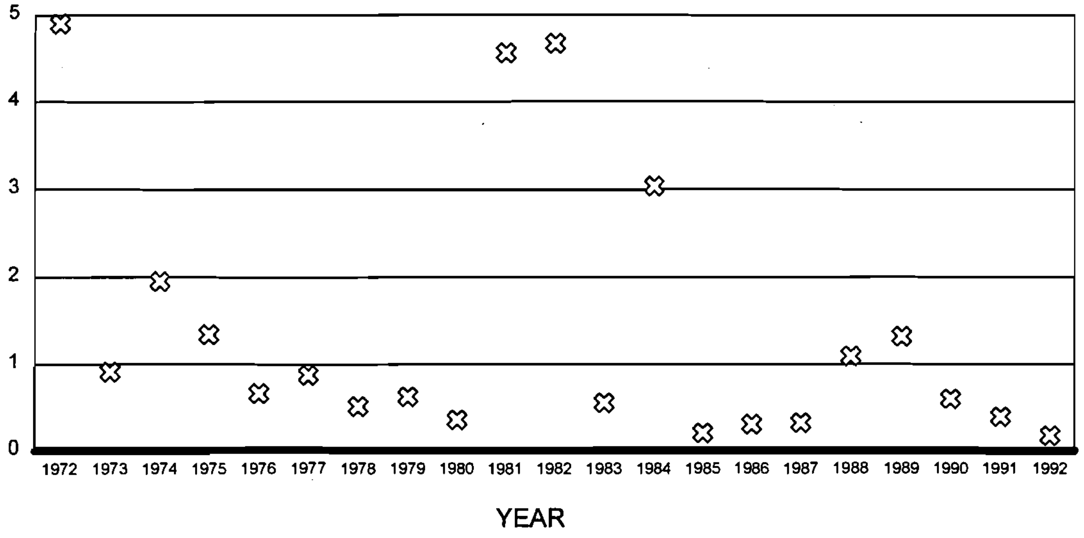
Figure 10 cont.

Reported Waterborne I-131 Emissions for All Lakes (in GBq)

Reactor Type: PWR



Date run: 12/22/97 GRAPH_1



Reported Airborne I-131 Emissions for All Lakes (in GBq)

Reactor Type: HWR

Date run: 12/22/97 GRAPH_1

Figure 10 cont.

Reported Airborne Cs-134 Emissions for All Lakes (in GBq)

Reactor Type: BWR

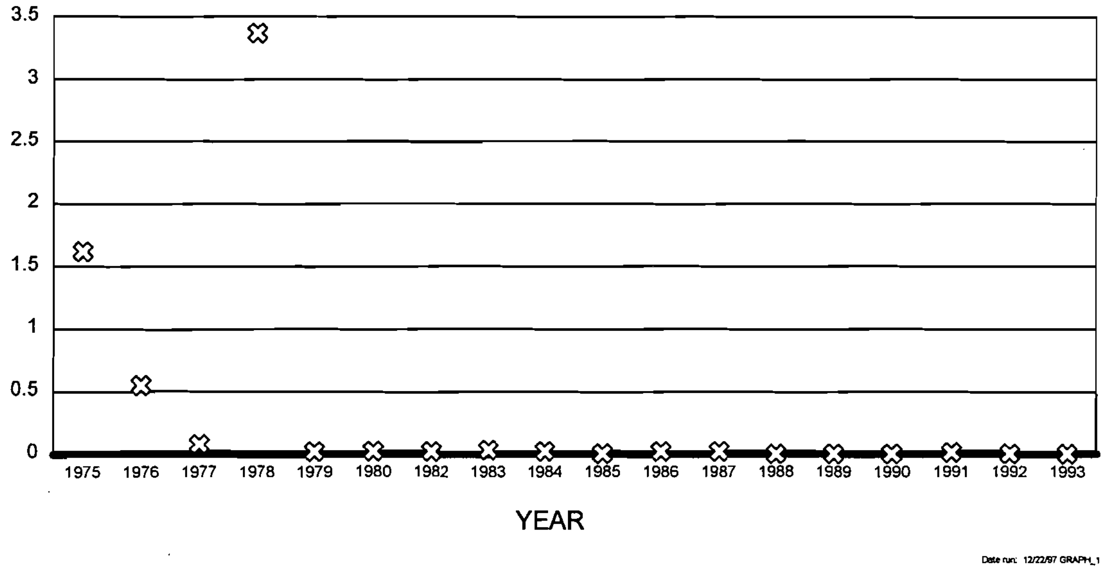
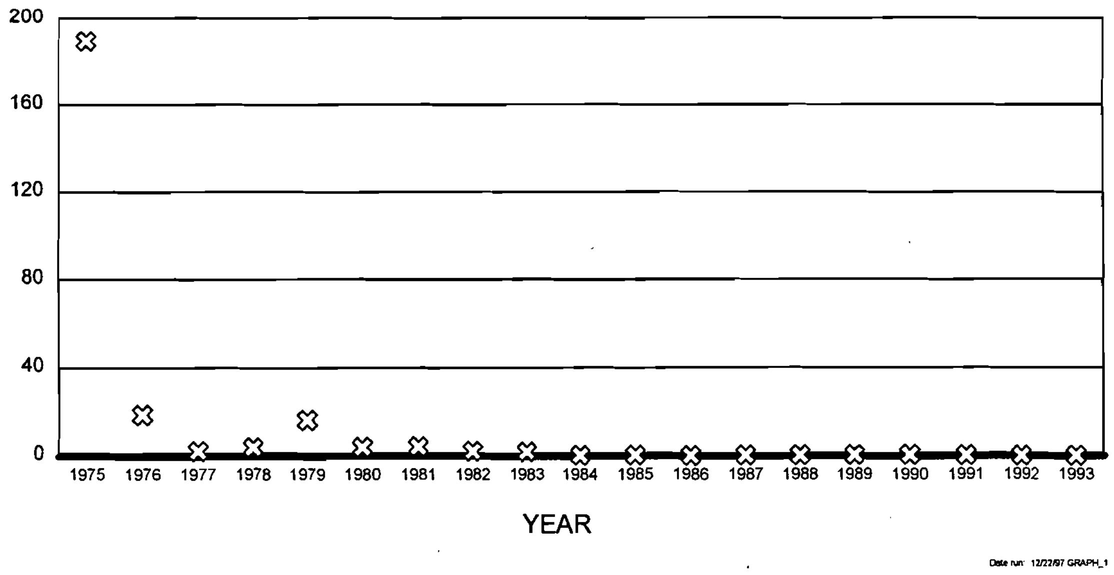
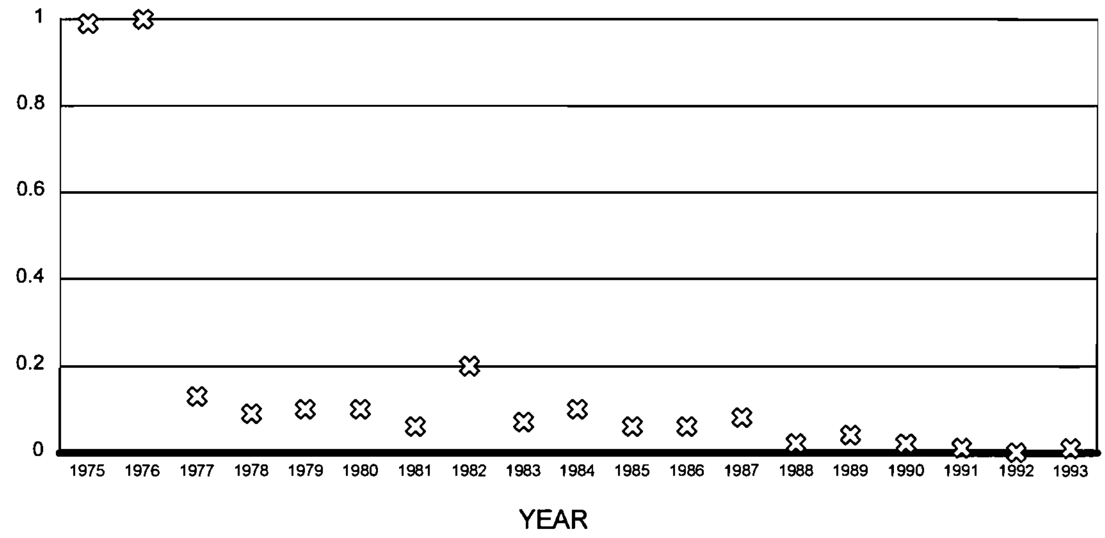


Figure 11

Reported Waterborne Cs-134 Emissions for All Lakes (in GBq)

Reactor Type: BWR



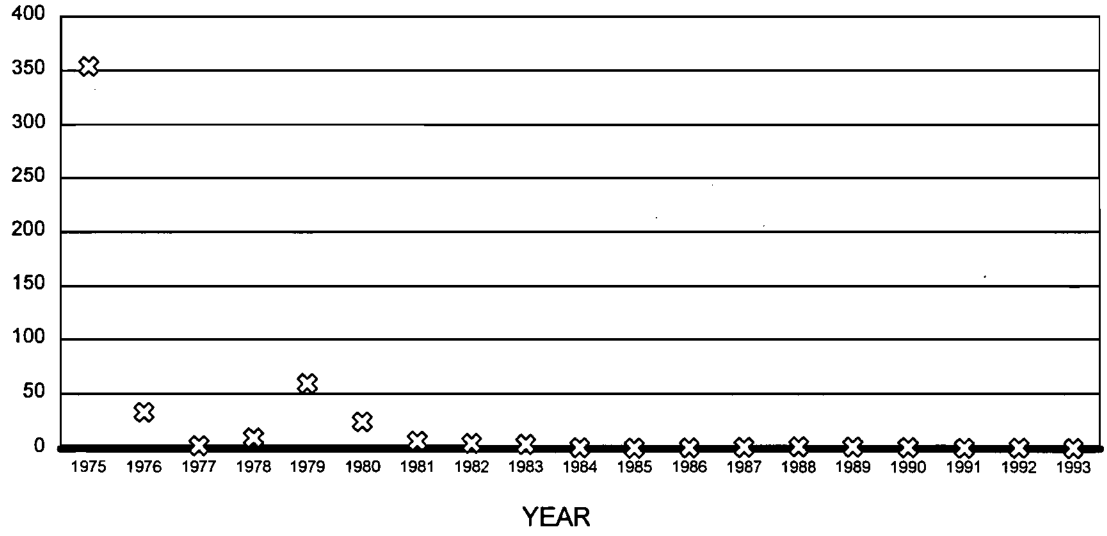


Reported Airborne Cs-137 Emissions for All Lakes (in GBq)

Reactor Type: BWR

Date run: 12/22/97 GRAPH_1

Figure 11 cont



Reported Waterborne Cs-137 Emissions for All Lakes (in GBq)

Reactor Type: BWR

Date run: 12/22/97 GRAPH_1

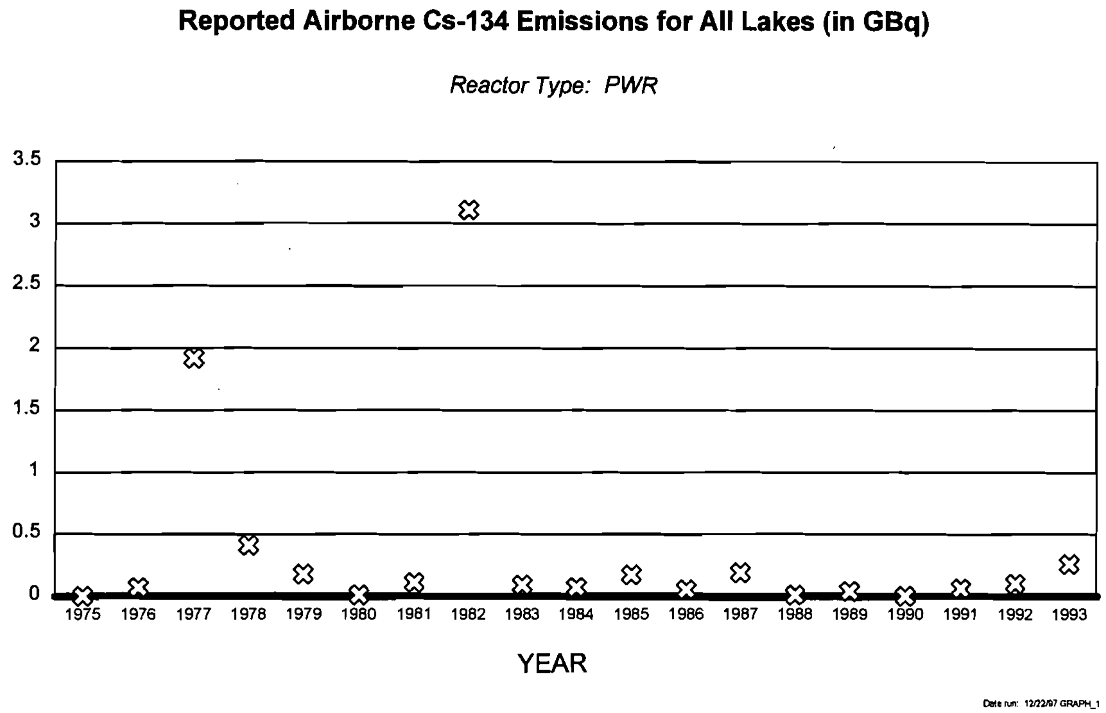
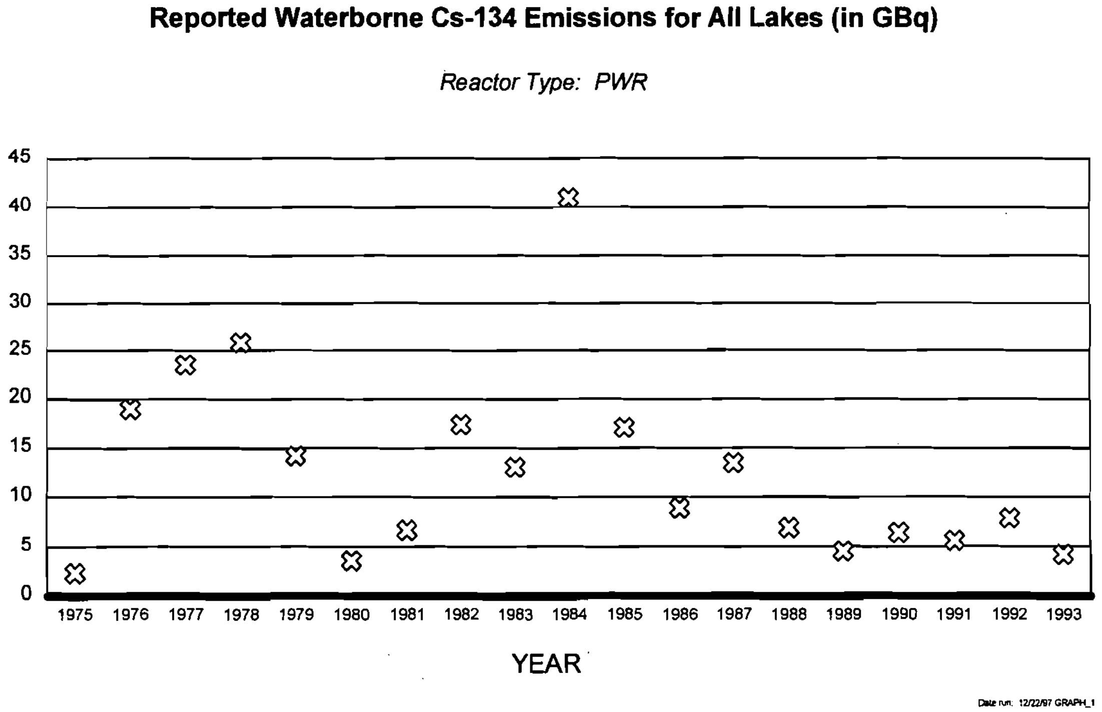

Reported Airborne Cs-134 Emissions for All Lakes (in GBq)
Reactor Type: PWR
3.5
3
2.5
2
1.5
1
0.5
0
1975
1976
1977
1978
1979
1980
1981
1982
1983
1984
1985
1986
1987
1988
1989
1990
1991
1992
1993
YEAR
Date run: 12/22/97 GRAPH_1

Figure 12


Reported Waterborne Cs-134 Emissions for All Lakes (in GBq)
Reactor Type: PWR
45
40
35
30
25
20
15
10
5
0
1975
1976
1977
1978
1979
1980
1981
1982
1983
1984
1985
1986
1987
1988
1989
1990
1991
1992
1993
YEAR
Date run: 12/22/97 GRAPH_1

Reported Airborne Cs-137 Emissions for All Lakes (in GBq)

Reactor Type: PWR

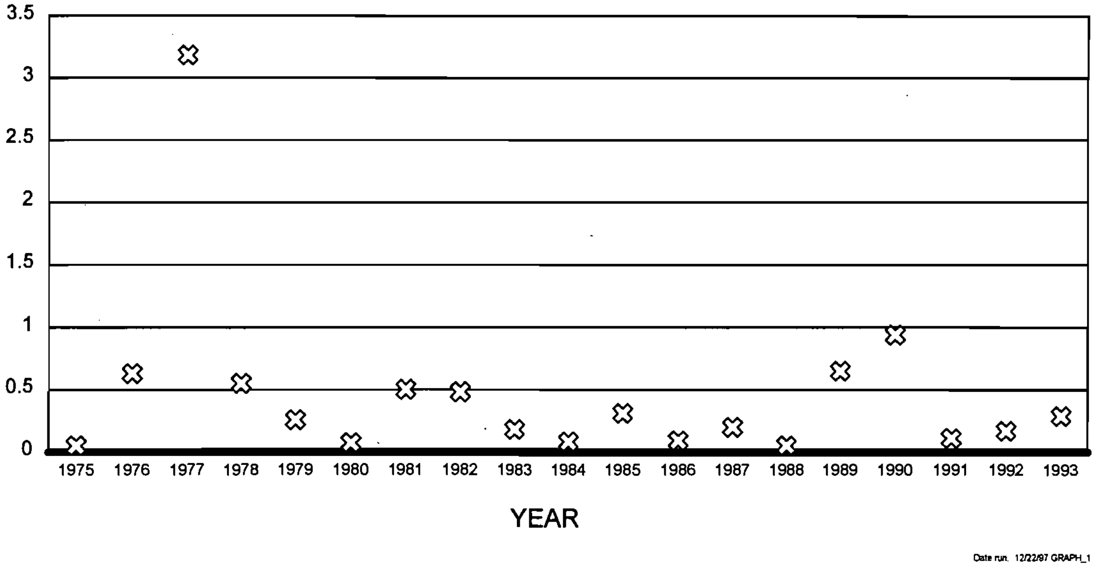
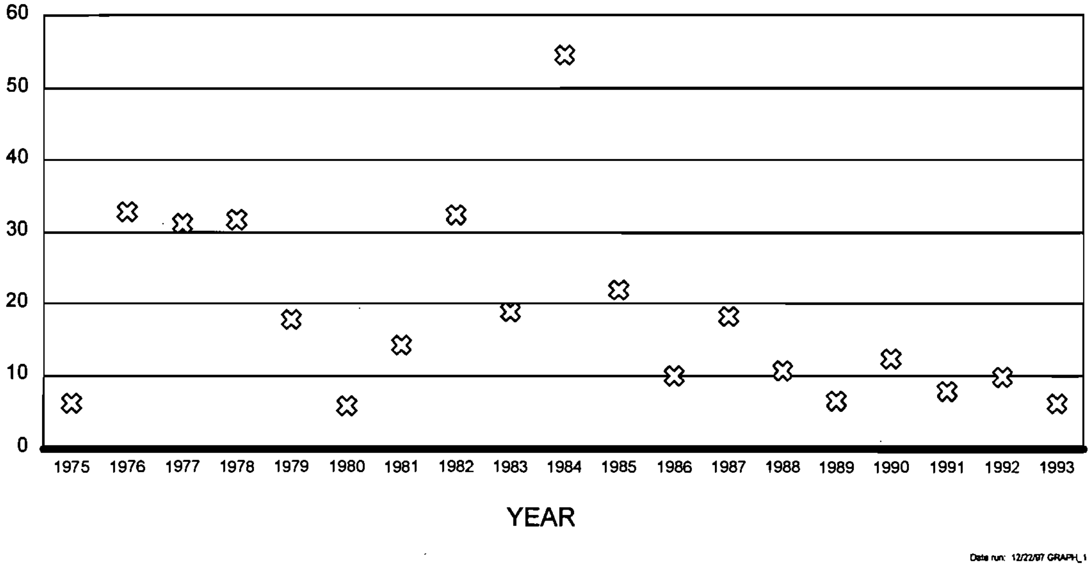
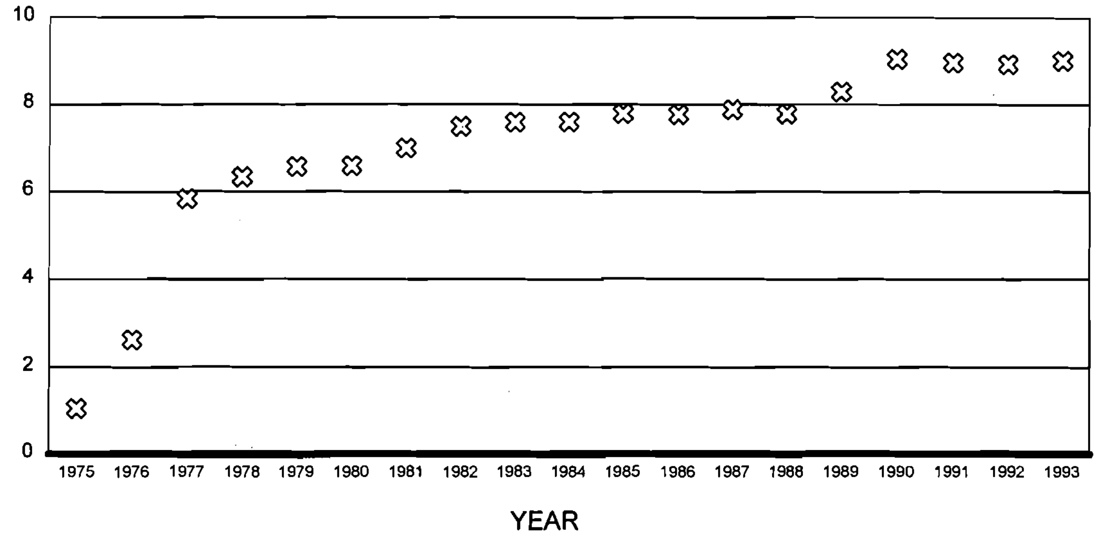


Figure 12 cont.

Reported Waterborne Cs-137 Emissions for All Lakes (in GBq)

Reactor Type: PWR

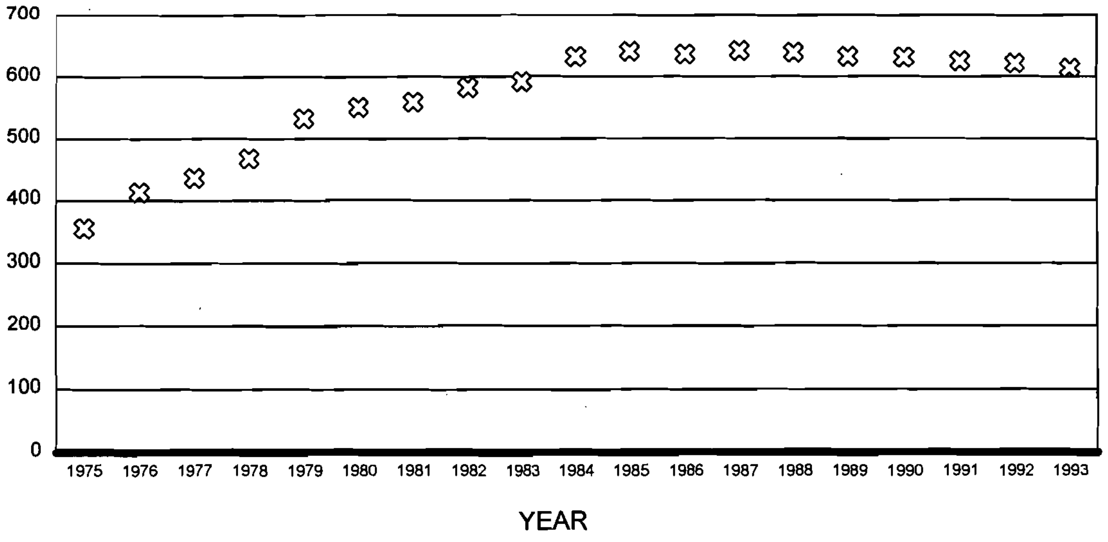



Reported Airborne Cs-137 Emissions for All Lakes (in GBq)

Cumulative Emissions corrected for Decay Reactor Type: ALL

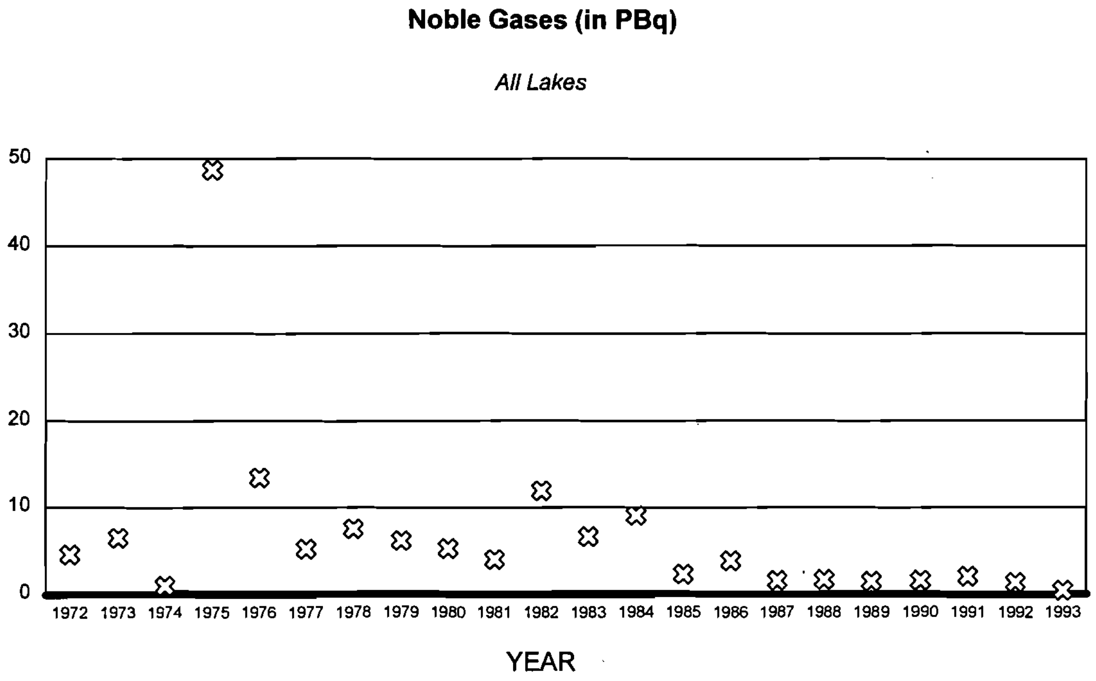
Date run: 12/22/97 GRAFDCAY

Figure 13


Reported Waterborne Cs-137 Emissions for All Lakes (in GBq)

Cumulative Emissions corrected for Decay Reactor Type: ALL

Date run: 12/22/97 GRAFDCAY



Date run: 12/22/97 GRAF_GAS

Figure 14

2.3.4 Emissions from Secondary Sources in the Great Lakes Basin

The sources not associated with releases from nuclear fuel cycle activities of radioactivity have been designated as "secondary sources." The terminology does not imply that the sources or their emissions are somehow secondary in importance. These non-nuclear fuel cycle sources of radioactivity to Great Lakes Basin are either military or civilian sources such as hospitals, industrial and commercial users, universities, or activities which release a "naturally occurring" radioactive material from an otherwise trapped matrix (technological enhancement), provided that the technological enhancement did not result from an activity associated with the nuclear fuel cycle. Although the emissions from a single source may be negligible, the large number of such sources in the Basin may make their combined effect significant. This discussion addresses open sources of radionuclides, which may eventually be released to the atmosphere or to the sewer systems draining into the lakes. We exclude an even larger category of sealed-source users, which would not be expected to release radionuclides to the air or water. The sealed sources could become a problem only if disposed of indiscriminately in municipal landfill sites.

All users of radioisotopes must obtain a license from the national regulator (the AECB in Canada or the Nuclear Regulatory Commission in the United States). Regular reporting of measured or estimated emissions is a condition of maintaining the licence. This information is available from the regulators; however, it is not usually in a format that is either convenient or machine readable. At the time of preparation of this report, the Task Force had obtained information from most of the Canadian users, but not on the larger number of United States users. Although these data are incomplete, we present them to give an indication of the magnitude of the emissions. One could obtain a crude estimate of total secondary emissions to the Basin by considering the ratio of the total population in the Basin to that on the Canadian side.

Hospitals and Universities

The Task Force inquiry on secondary sources emphasized research reactors at universities and industrial sites and the use of radioisotopes in hospitals, research facilities, and medical facilities other than hospitals. The Canadian data, obtained with the cooperation of the AECB, came through questionnaires sent to all its licensees asking them to estimate their emissions. Responses were obtained from 45% of the licensees, which was assumed to include virtually all the licensees with significant emissions. The users of the greatest quantities of radionuclides are nuclear medicine departments of hospitals, which administer radioisotopes to patients for diagnostic purposes. Lesser amounts of radionuclides are used for research or industrial purposes. Generally, about 75% of the radioisotopes administered to patients are assumed to be excreted to sewers.

Table 16 summarizes the results for the secondary Canadian users of radioisotopes for the years 1993, 1994, and 1995. For most radionuclides the emissions are a few megabecquerels per year, but a few can reach the gigabecquerel per year levels. These levels are insignificant compared with the terabecquerel and petabecquerel levels released from nuclear reactors. Also, the radionuclides from secondary sources all have half-lives much less than one year and therefore do not accumulate from year to year.

TABLE 16
EMISSIONS FROM SECONDARY SOURCES IN CANADA TO THE GREAT LAKES BASIN BY RADIONUCLIDE

			Activity (MBq)		
Radionuclide	Half-Life (days)	Type	1993	1994	1995
^{45}Ca	163	Patients	40	8.9	2.7
		Research	67	0.30	0.38
		Sewer		140	360
		Total	107	149.2	363.1
^{57}Co	271	Patients	260	280	310
		Sewer	23	41	38
		Total	283	321	348
^{58}Co	70.8	Patients	93	93	93
		Sewer	0.37	0.37	0.37
		Total	93.4	93.4	93.4
^{32}P	14.3	Patients	7100	6200	7400
		Research		0.088	0.14
		Sewer	8900	9600	9900
		Total	16,000	15,800	17,300
^{33}P	25.3	Sewer	42	56	400
		Total	42	56	400
^{35}S	87.2	Research		0.51	0.43
		Sewer	12,000	11,000	16,000
		Total	12,000	11,000	16,000
^{51}Cr	27.7	Patients	2600	2500	2000
		Research		0.054	
		Sewer	1600	1000	940
		Total	4200	3500	2940
^{125}I	59.7	Patients	16,000	8400	6800
		Research		2.9	3.0
		Sewer	3400	3400	3900
		Total	19,400	11,803	10,703
^{131}I	8.04	Patients	2.6×10^6	2.9×10^6	3.0×10^6
		Sewer	0.71×10^6	0.73×10^6	0.65×10^6
		Total	3.31×10^6	3.63×10^6	3.65×10^6

3 INVENTORY BY GEOGRAPHICAL DISTRIBUTION OF RADIOACTIVITY IN THE GREAT LAKES AIRSHED AND WATERSHED

3.1 The Whole Lake Data

3.1.1 Geographical Distribution of Radionuclides

Because of the many different sources of radioactivity to the Great Lakes, their patterns of release and the actions of various environmental processes, the geographical distribution of radionuclides in the Great Lakes shows considerable irregularity and non-uniformity. The presence of a specific radioactive isotope in one of the Lakes does not assure its presence in the other Lakes or connecting channels. Even within a lake, the distribution is not uniform (does not suggest a totally mixed lake) but often shows a stratification with a different activity in the nearshore region compared with the larger open lake region. Thus, some comments about geographical distribution of radionuclides are essential.

Theoretically, all radioisotopes found in atmospheric fallout, regardless of origin (cosmogenic or as the result of past weapons testing or accidents such as at Chernobyl), should appear in the waters and sediments of all of the Great Lakes and connecting channels. Only a very few long-lived isotopes, however, are detectable either in dry or wet precipitation to the Basin. Also, these isotopes may not be detected in all of the air, water, or sediment samples from all of the Great Lakes. Since rates of atmospheric deposition depend mainly on the surface area for the deposition, Lake Superior, with the greatest surface area of the Great Lakes would receive the highest load of isotopes from deposition. However, Lake Superior has no nuclear power plant facilities discharging to it, suggesting that the only isotopes expected from atmospheric deposition would be those associated with past nuclear weapons testing or originate from sources a considerable distance away from the Great Lakes and have been subject to long-range atmospheric transport.

The Task Force used the work of the Commission's International Air Quality Advisory Board to examine the typical atmospheric transport of materials from various locations within North America. Prevailing wind patterns to the Great Lakes from North American sources would suggest the possibility that the Hanford facility (Richland, Washington) and Idaho Falls facility of the Department of Energy might be suitable sources of atmospheric transport of radioactive materials to the Great Lakes. However, the monitoring reports of both facilities, which include high-altitude samples, did not show any detectable radionuclides known to be discharged in air emissions from those facilities reaching the Great Lakes within 5 days, which is sufficient time to transport more than 90% of the materials with half-lives greater than 5 days.

3.1.2 Environmental Monitoring Data from Nuclear Reactor Facilities

The Nuclear Task Force has collected and examined environmental monitoring data provided by the operators of the major nuclear facilities in the Great Lakes Basin. Virtually all of the reported radionuclides had activities or concentrations that were reported as the lower limit of detection (LLD). This does not necessarily mean that the various radionuclides were absent from the environment, nor that their environmental impacts were insignificant. It simply means that the radionuclides could not be detected by the instrumentation and procedures used.

The LLD for a measurement depends on several factors including sensitivity of the instrumentation, length of time that radioactivity from the sample is counted, and elapsed time between sample collection and counting, (a shorter elapsed time means fewer losses due to radioactive decay). Facility operators must often seek a compromise between the need for a low level of detectability, the costs of the instrumentation, and the need for a large throughput of samples in a limited amount of time. Operating parameters are often chosen for the purpose of demonstrating compliance with government regulations rather than characterizing the movement of radionuclides in the environment.

Rather than attempting to reproduce the results from all of the station reports, we have chosen three typical examples to illustrate the general significance and the limitations of these results. Table 17 shows the results for artificial radionuclides in shoreline sediments, fish, and surface waters near the Nine Mile Point Nuclear Station (Oswego County, New York) during 1994. ^{137}Cs was the only artificial radionuclide detected in sediments and fish, and tritium (^{3}H) was the only one detected in surface water. For the fish and water, there were no significant differences in the measured levels between the indicator locations (column 4) and the control locations (column 6). This would indicate that the measured levels were not due to emissions from the local facilities. Only the ^{137}Cs in sediment appeared to be elevated at the indicator locations, suggesting a contribution from the power station.

Tables 18 and 19 show environmental monitoring results from the Donald G. Cook Nuclear Plant operated by the Indiana Michigan Power Company. In Table 18, ^{131}I levels in air were above the limit of detection for a brief period from February 21 to March 14, 1994. These were directly attributed to station emissions. Table 19 shows that, apart from the naturally occurring ^{40}K (potassium-40), the only radionuclide detected in fish during 1994 was ^{137}Cs. These levels are typical of global fallout and do not appear to have been enhanced by station emissions.

In Tables 17–19, the original units of picocuries (pCi) have been retained to illustrate how lower limits of detection are reported (1 pCi = 0.037 Bq).

Table 20 shows levels of radionuclides in Lake Ontario fish in the vicinity of the Pickering Nuclear Generating Station during one year (1988). The level of naturally occurring potassium-40 in fish and other biota is regulated by a homeostatic mechanism and is unaffected by inputs from human activities. The ^{14}C and ^{137}Cs detected in the fish are probably due to residual fallout from the earlier testing of nuclear weapons. Levels of 250–350 Bq of ^{14}C per kg carbon were typical of background values in 1988. One to 2 Bq/kg of ^{137}Cs are quite typical of fish taken from lakes across Canada (Elliott et al. 1981).

TABLE 17
RADIOLOGICAL ENVIRONMENTAL MONITORING PROGRAM ANNUAL SUMMARY
NINE MILE POINT NUCLEAR STATION UNIT 1 DOCKET NO. 50-220
NINE MILE POINT NUCLEAR STATION UNIT 2 DOCKET NO. 50-410
OSWEGO COUNTY, STATE OF NEW YORK, JANUARY – DECEMBER 1994

Medium (units)	Type and number of analyses	LLD	Indicator locations: mean, frequency, and range	Highest annual mean: mean, frequency, range, and location [a]		Control location: mean, frequency, and range
Shoreline Sediment (pCi/kg-dry)	^{134}Cs	150	<LLD	<LLD		<LLD
	^{137}Cs	180	237 (2/2)[b] 84–390	237 (2/2) 84–390	Sunset Bay: 1.5 at 80^0	<LLD
Fish (pCi/kg-wet)	^{54}Mn	130	<LLD	<LLD		<LLD
	^{59}Fe	260	<LLD	<LLD		<LLD
	^{58}Co	130	<LLD	<LLD		<LLD
	^{60}Co	130	<LLD	<LLD		<LLD
	^{65}Zn	260	<LLD	<LLD		<LLD
	^{134}C	130	<LLD	<LLD		<LLD
	^{137}Cs	150	20 (7/19) 11–28	24 (4/11) 12–35	OSS: 7.6 at 235°	24 (4/11) 12–35
Surface Water (pCi/L)	^{3}H	3000	220 (2/4) 180–260	250 (1/4) 250–250	OSS: 7.6 at 235°	250 (1/4) 250–250
	^{54}Mn	15	<LLD	<LLD		<LLD
	^{59}Fe	30	<LLD	<LLD		<LLD
	^{58}Co	15	<LLD	<LLD		<LLD
	^{60}Co	15	<LLD	<LLD		<LLD
	^{65}Zn	30	<LLD	<LLD		<LLD
	^{95}Zr	15	<LLD	<LLD		<LLD
	^{95}Nb	15	<LLD	<LLD		<LLD
	^{131}I	15	<LLD	<LLD		<LLD
	^{134}Cs	15	<LLD	<LLD		<LLD
	^{137}Cs-137	18	<LLD	<LLD		<LLD
	^{140}Ba/^{140}La	15	<LLD	<LLD		<LLD

Notes: (a) Location is distance in miles and direction in compass degrees based on NMP-2 reactor centerline.

(b) Underlined values are the mean (237), with frequency (ratio of number of detectable measurements to total number of measurements) given in parentheses(2/2), and the range of values given underneath (84–390).

TABLE 18
INDIANA MICHIGAN POWER COMPANY — DONALD C. COOK NUCLEAR PLANT
IODINE-131 IN WEEKLY AIR CARTRIDGE SAMPLES
Results in Units of 10^{-3} pCi/m^3 ± 2 SD

Collection dates	Station codes									
	A-1	A-2	A-3	A-4	A-5	A-6	Coloma	Dowagiac	New Buffalo	South Bend
January 1994										
01/03/94	<10	<10	<20	<10	<7	<8	<8	<9	<6	<8
01/10/94	<10	<10	<10	<10	<8	<20	<20	<20	<8	<20
01/17/94	<20	<20	<10	<20	<10	<20	<20	<20	<10	<2
01/24/94	<20	<20	<20	<10	<7	<8	<8	<8	<6	<8
01/31/94	<10	<10	<10	<10	<7	<20	<20	<20	<10	<20
February 1994										
02/07/94	<20	<20	<20	<20	<7	<9	<8	<8	<6	<8
02/14/94	<10	<10[a]	<10	<10	<8	<10	<10	<10	<8	<10
02/2/94	<20	<9[a]	<20	<20	<7	<10	<20	<20	<10	<10
02/28/94	<7	<5[a]	<7	10.1 ± 5.2	<10	<20	<20	<20	<8	<20
March 1994										
03/07/94	<10	21.0 ± 10.4	<10	<10	26.7 ± 8.4	<20				<10
03/14/94	8.8 ± 5.0	<9	<8	11.2 ± 5.7	<10	<10				<10
03/21/94	<10	<20	<10	<10	<10	<10				<10
03/28/94	<20	<20	<20	<20	<30[b]	<10				<10

Notes: (a) No measurable volume. Results in total pCi.
(b) Equipment malfunction; low sample volume.
SD, Standard deviation.

TABLE 19
INDIANA MICHIGAN POWER COMPANY — DONALD C. COOK NUCLEAR PLANT
CONCENTRATIONS OF GAMMA EMITTERS IN FISH
Results in units of pCi/kg wet weight ± 2 SD

Collection Date	Station	Description	^{7}Be	^{40}K	^{137}Cs	^{228}Ra	^{228}Th
06/08/94	OFS-South	Lake trout	<90	2980 ± 300	<10	<200	<10
06/08/94	ONS-South	Lake trout	<100	3030 ± 300	<20	<200	<20
06/08/94	ONS-North	Lake trout	<100	2970 ± 300	<20	<200	<20
06/08/94	OFS-North	Sucker	<100	2490 ± 250	19.5 ± 10.0	<200	<20
10/18/94	OFS-South[a]						
10/18/94	ONS-South	Walleye	<100	3420 ± 340	59–60 ± 14.3	<300	<30
10/18/94	ONS-North	Sucker	<100	3500 ± 350	80.1 ± 14.0	<300	<20
10/18/94	OFS-North	Sucker	<100	2870 ± 290	28.8 ± 11.9	<300	<30
Average ± 2 SD				3039 ± 685	47.0 ± 55.9		

Notes: (a). Sample unavailable.
SD, Standard deviation.

TABLE 20
LAKE ONTARIO FISH
PICKERING NUCLEAR GENERATING STATION AREA

Location	Species (Composites)	Sample Date	^{14}C** (Bq/kg-C, ±2SD)	Gamma (Bq/kg, ±2SD)* ^{137}Cs	^{134}Cs	^{40}K
"A"	Brown Trout	15 Aug. 88	280 ± 40	0.97 ± 0.16	<0.13	108 ± 5
Discharge	Lake Trout	15 Aug. 88	280 ± 40	1.82 ± 0.26	<0.18	169 ± 7
	Sucker	15 Aug. 88	370 ± 50	0.46 ± 0.15	<0.13	122 ± 5
"B"	Lake Trout	01 Oct. 88	270 ± 40	2.00 ± 0.27	<0.17	141 ± 6
Discharge	Brown Trout	15 Aug. 88	270 ± 40	0.96 ± 0.17	<0.12	125 ± 5
	Sucker	15 Aug. 88	330 ± 40	0.88 ± 0.22	<0.19	186 ± 7
Duffin's	Lake Trout	15 Aug. 88	220 ± 40	2.09 ± 0.27	<0.19	193 ± 8
Creek	Brown Trout	15 Aug. 88	340 ± 40	1.14 ± 0.19	<0.14	145 ± 6
	Sucker	15 Aug. 88	460 ± 70	0.90 ± 0.22	<0.20	180 ± 7

Notes: 1 Bq = 27 pCi

* Bq/kg - wet weight, edible fish flesh. The ± values represent twice the standard deviation of one determination.

** The ± values represent twice the standard deviation of the mean of two determinations.

Discharges and runoff from the West Valley waste storage site enter Buttermilk Creek, which discharges into Cattaraugus Creek, which then discharges into Lake Erie. Cattaraugus Creek has received continuous radiological monitoring since 1968. Small amounts of radioactivity are detected in the creek, and water samples collected below the discharge area of the facility contain measurable levels of radioactivity. Data through 1985 were previously reported by the Commission (1987*b*). More recent data through 1993 were graciously supplied by Mr. William Condon of the New York State Department of Health's Bureau of Environmental Protection (N.Y. Department of Health 1983–1993). Table 21 summarizes data on the monitoring of Cattaraugus Creek.

TABLE 21
RADIONUCLIDE LOADINGS TO THE GREAT LAKES FROM CATTARAUGUS CREEK AND THE WEST VALLEY NUCLEAR SERVICE CENTRE SITE

Year	Annual Average Concentration (pCi/L)			
	Gross α	Gross β	^{90}Sr	^{3}H
1968	—	123	25	22,000
1969	—	214	47	17,600
1970	—	222	69	19,600
1971	ND	208	37	31,000
1972	ND	169	9	2200
1973	ND	19	4	<500
1974	<4	15	<3	<800
1975	<4	11	<3	6200
1976	<5	10	<2	8400
1977	<5	7	1	1800
1978	<4	7	1	2700
1979	<3	5	1	350
1983	<1.3 – <7	<1.4 – 8.0	<0.6 – 1.4	<130 – 330
1984	<3.0 – 16	8.2	1	<150 – 300
1985	<2.0 – 9.0	<3.0 – 10.0	<0.6 – 1.3	<130 – 500
1986	<2	6	1	500
1987	<3	6	1	500
1988	<3	5	1	500
1989	<3	5	1	500
1990	<3	12	1	500
1991	<3	4	1	500
1992	<4	6	1	<200
1993	<4	4	1	<200

Notes: (1) Station Location: Springville Dam (Site 1459-042)

(2) Data sources: International Joint Commission (1983), International Joint Commission (1987*b*), New York State Department of Health (Mr. William Condon, personal communication Apr. 12, 1996)

(3) Symbols: (—) not measured or not reported; (<) less than lower limit of detection of instrumentation in use (LLD) or offscale in some manner; (ND): not detected.

(4) U.S. EPA Drinking Water standards: If gross β exceeds 50 pCi/L, then analysis for specific radionuclides must be performed. The allowable upper limit of gross β radiation is 50 pCi/L. The allowable upper limit for gross α radiation is 15 pCi/L of which the combined activities of the radionuclides ^{226}Ra and ^{228}Ra cannot exceed 5 pCi/L.

3.1.3 Water-Column Inventories for the Open Lakes

A simple material balance for a radionuclide in a lake has three source terms (atmospheric fallout, tributary inflow, and direct discharge of effluents to a lake), two storage terms (water column and sediments), and an export term (lake outflow). A sixth term, also an export term, the revolatilization of gaseous radionuclides from a lake surface, applies to a very few radionuclides, mainly those of the noble gases (*e.g.*, ^{41}Ar, ^{133}Xe, ^{85}Kr, ^{222}Rn), ^{14}C in radioactive methane and carbon dioxide, and tritium in water. While tritium moves through the environment via the hydrological cycle, volatilization from the Lakes does not appear in any of the inventory calculations available for this radionuclide.

By considering the Great Lakes as a system of five lakes and several connecting channels, material balances are possible at two levels of scale: an individual lake, and the "whole" system of lakes and connecting channels. The hydrological parameters of the Great Lakes are shown in Table 22. The material balances share many terms, and the system has several simplifying features. The absence of nuclear power plants on Lake Superior or its tributaries means the input of radioactivity to this Lake comes mainly from atmospheric deposition. The outflows from Lake Superior become inflows to Lakes Michigan and Huron, and the outflows from these lakes become inflows to connecting channels to Lake Erie, whose outflow is Lake Ontario's inflow, and Lake Ontario's outflow goes to the St. Lawrence River.

In a relatively uniform (completely mixed) lake, the product of the lake volume and its average radionuclide concentration or activity estimates the inventory term for lake water storage of the radionuclide. Many inventory calculations separate the inventories of water column and sediments but fail to report the sediments. See the section, **Sediments**, below for further discussion of sediment inventories.

The inventory of radionuclides in the water column is often difficult, because radionuclides are difficult to detect unless very large samples of water (300 L) are taken. The major exceptions are tritium and isotopes of strontium and rubidium, which because of water solubility of most of their inorganic compounds, remain in the water column preferentially over the sediments.

The long history of monitoring of ^{90}Sr and ^{137}Cs (each with half-lives of about 30 years) is the basis for calculation of the deposition inventories of many other radionuclides. The calculation depends on using geochemical ratios with latitude band adjustments and begins with choosing a given year and noting the depositional flux for ^{90}Sr or ^{137}Cs from the worldwide fallout data for latitude bands of 30–40°N and 40–50°N (which cover the area of the Great Lakes). The longest fallout record is for ^{90}Sr data with ^{137}Cs fluxes estimated using a ratio of the activities of ^{137}Cs/^{90}Sr of 1.6. This ratio describes a relatively constant distribution of the two radionuclides in many environmental media and compartments.

For other radionuclides, the activity ratio of a radionuclide to either the strontium or cesium in fallout is used. Data from the atmospheric testing of nuclear weapons during the period of 1954–1963 provided usable ratios. Wahlgren et al. (1980) used 0.176 for the activity ratio ^{239,240}Pu/^{90}Sr to estimate the atmospheric deposition of plutonium to the Great Lakes. Separate data sets exist for ^{210}Pb, which has been intensively studied as an atmospheric tracer. This isotope has both natural and artificial sources and, therefore, was not studied by the Task Force.

TABLE 22
HYDROLOGICAL PARAMETERS FOR THE GREAT LAKES

Lake	Drainage basin (10^4 km^2)	Lake surface (10^4 km^2)	Volume (10^3 km^3)	Outflow (km^3/ year)	Depth (m)
Superior	12.8	8.24	12	65	149
Michigan	11.8	5.8	4.9	49	84
Huron	13.1	5.96	4.6	154	77
Erie	5.9	2.57	0.48	175	17
Ontario	6	1.95	1.6	209	86

Joshi (1991) has noted that, except for ^{3}H, ^{90}Sr, ^{137}Cs, and ^{239,240}Pu, data for very few radionuclides have been reported for open lakes. Some open lake data are available for uranium, radium, and thorium in Lakes Huron, Michigan, and Ontario, but most inventory calculations require geochemical correlation and estimation methods. Both Joshi and the GLWQB (IJC 1983, 1987*b*) have noted that radionuclide data reported for drinking water intakes often do not differ statistically from open lake cruise data, and that averaging the data on a radionuclide from drinking water intakes at different locations in a lake may provide a reasonable estimate of the open lake or even whole-lake average radionuclide concentrations.

Although the Task Force has examined a significant quantity of environmental data collected by nuclear facility operators, the analysis of open water data was constrained by the limited number of lakewide monitoring surveys conducted in the past, making environmental and biological assessments difficult to perform. The Surveillance Work Group of the Commission's Water Quality Board (IJC 1987*b*) has previously recommended that radionuclide monitoring be conducted in the open waters of the Great Lakes every 5 years in a manner similar to the surveys conducted by the National Water Research Institute, Environment Canada, between 1973 and 1983. The last open water surveillance program was initiated in 1990 by Environment Canada, Conservation and Protection, but was limited to Lake Ontario. Its scope was to ensure that nuclear facilities and other sources of radioactive contamination were controlled in a manner that met the broad objectives on the Lake Ontario Toxics Management Plan and the Great Lakes Water Quality Agreement (IJC 1987*a*).

Table 23 gives the average concentrations in open lake water of the three major radionuclides (^{3}H, ^{90}Sr, and ^{137}Cs), together with their estimated inventories in the water column. Also shown are the estimated inputs to 1983 from fallout and to 1993 from reactors. Virtually all of the fallout deposition was complete by 1983. No attempt has been made here to carry out a mass balance. It must be recognized that large quantities of radionuclides have been lost to the water column through outflow and sedimentation.

TABLE 23
INVENTORIES OF TRITIUM, STRONTIUM-90, AND CESIUM-137 IN THE GREAT LAKES

	Concentration (Bq/L)[1] in 1983	Inventory in 1983 (TBq)	Fallout input to 1983 (TBq)[1,3]	Reactor input to 1993 (TBq)
^{3}H				
Superior	6.7	80,000	70,000	No reactors
Michigan	7.4	36,000	60,000	1300
Huron	10.6	49,000	70,000	16,100
Erie	8.5	4100	40,000	70
Ontario	13.5 (10.1)[2]	22,000	30,000	11,000
All lakes		191,100	270,000	28,500
^{90}Sr				
Superior	0.016	192	123	No reactors
Michigan	0.017	83	98	0.007
Huron	0.023	106	98	No data
Erie	0.024	12	45	<0.001
Ontario	0.025 (0.024)[2]	40	33	0.003
All lakes		433	397	0.01
^{137}Cs				
Superior	0.0017	20	200	No reactors
Michigan	0.0012	5.9	159	0.35
Huron	0.0012	5.5	159	No data
Erie	0.0006	0.29	74	<0.01
Ontario	0.0008	1.3	54	0.51
All lakes		33	646	0.86

Notes: (1) Joshi (1991).
(2) Environment Canada, 1990 Lake Ontario Survey.
(3) Ahier and Tracy (1995).

Radionuclide levels measured during the 1990 Lake Ontario survey are similar to those from the 1983 survey. ^{3}H in open waters ranged from about 9.1 to 10.8 Bq/L (average: 10.1 Bq/L); concentrations at sites near the inflow to Lake Ontario averaged about 6.8 Bq/L, and a level of 9.2 Bq/L was measured at the outflow (Environment Canada, unpublished data). More recent sampling of ^{3}H levels in Lake Ontario between 1991–1993 showed average concentrations between 9 and 11 Bq/L, with a projected yearly increase due to routine CANDU operations of about 0.12 Bq/L (Chant et al. 1993). Other radionuclides measured in Lake Ontario open waters in 1990 include uranium (0.270 µg/L), ^{226}Ra (0.002 Bq/L), and ^{210}Pb (0.12 Bq/L). Levels of ^{90}Sr in fish ranged from <0.01–0.059 Bq/g in flesh and 0.018–0.059 Bq/g in bone. ^{137}Cesium in fish flesh ranged from <0.01 to 0.064 Bq/g; all levels measured in bone were <0.01 Bq/g.

The inventory for plutonium is a model for further calculations as it is based on actual field data with only the atmospheric deposition term estimated from ^{90}Sr. Table 24 presents the inventory of plutonium from the work of Wahlgren et al. (1980). The Task Force considers that inventory one of the most effective uses of field data, extrapolation, and modeling methods of any of the radionuclide inventories in the literature.

Field studies of plutonium rarely distinguish between the two isotopes, ^{239}Pu and ^{240}Pu, but rather tend to report plutonium as a sum of all isotopes. Some limited data suggest a typical isotope distribution of 60% ^{239}Pu, 30% ^{240}Pu, and the remaining 10% divided among short-lived plutonium isotopes and decay chain radionuclides, but that does not seem to compromise the usefulness of the data.

TABLE 24
PLUTONIUM INVENTORY (TBq) FOR THE GREAT LAKES
(adapted from Wahlgren et al. 1980)

Lake	Watershed	Deposited on Surface	Water Column	Sediments
Superior	11	6.7	0.2	6.5
Michigan	10	4.8	0.07	4.7
Huron	10	4.8	0.07	4.7
Erie	7.0	3.0	0.04	3.1
Ontario	5.2	1.5	0.04	1.8

Notes: (1) Estimates were for 1977.
(2) The surface deposition calculation is based on using ^{90}Sr data with a $^{239,240}Pu/^{90}Sr$ geochemical ratio of 0.0176.
(3) Plutonium isotopes occur mainly as insoluble oxides and move to the sediments. Revolatilization is not an important process.

3.1.4 Sediments

As noted, most radionuclides entering the Great Lakes move to sediments as their final repositories. The separation of radionuclide fractions between water column and sediments depends on a distribution parameter, K_D (defined as the ratio of the activity of a radionuclide per unit weight of sediment to the activity of that same radionuclide per unit weight of the bulk water phase or water column), which quantifies the distribution of activities of a radionuclide in a bulk sediment and bulk water column. Edgington has pointed out to the Task Force that a K_D of less than 1,000 often means that there is a detectable fraction of the activity of a radionuclide in the water column, a K_D between 1,000 and 10,000 often means that the distribution of radioactivity between sediment and water column is unpredictable, and K_D of greater than 10,000 assures that the radionuclide is almost totally incorporated into the sediment (greater than 99.99%). Although the parameter, K_D, is empirical, its use has some theoretical basis in surface chemistry. Values of K_D depend on sediment properties: consolidation, pore water, strength of bottom currents that cause scour, the chemical environment (*i.e.*, alkalinity, pH, Eh, ionic strength, etc.), and the biological activity of benthic organisms. In undisturbed sediments, radionuclides that bind to particulates or bottom materials tend to remain in the upper few centimeters of surficial sediments.

3.2 Inventories for Biological Compartments

One of the most difficult components of an inventory is the assessment of the radionuclide content of the biota. Organisms are continuously exposed to radiation and radioactivity, but the extent to which they act as repositories for radioactive isotopes of various elements involves a complex set of metabolic and physiological processes, which have not been intensively studied for purposes of establishing an inventory. Most of the research entails the use of radioactive versions of selected elements or compounds (tracers), which are important in the physiological functioning of various species in order to understand the pathways and mechanisms of those physiological processes and functions. Almost none of the studies extended the data from tracer studies to establish biological compartmental inventories of radionuclides.

3.2.1 Bioaccumulation and Biomagnification

This section addresses individual elements and radionuclides with respect to bioaccumulation and biomagnification factors for freshwater biota. Bioaccumulation refers to the retention in a biological compartment of material from the external environment or a *non-biological* or *non-living* source material to an extent or in an amount that exceeds on a relative weight basis the presence of the material in the source. Biomagnification refers to the retention in a biological compartment of a material that originated from *another biological compartment* in an amount that exceeds on a relative weight basis its presence in the originating biological compartment. Thus, bioaccumulation applies to the uptake of materials from water and sediments as non-living source materials, and biomagnification applies to the uptake of materials from living source materials, as in predator–prey interactions. The quantification of bioaccumulation and biomagnification is through biological transfer factors, which are defined for radionuclides in a manner identical to the definition of the K_D of sediments:

> a *ratio of activity of a given radionuclide on per unit weight basis for biological tissue to non-living source material, and a ratio of activity of a given radionuclide on per unit weight basis for a biological repository to that of its biological (living) source material.*

The work emphasizes studies with stable radionuclides, but some data derived from radionuclides appear, mainly radionuclides of cesium and potassium. Table 25 lists those radionuclides in increasing order of atomic weight and number, which because of their half-lives, could be of possible interest with respect to the limnological cycling of elements through the biota of the Great Lakes. Several radionuclides appear as "combinations," notably ^{140}Ba/^{140}La, ^{95}Zr/^{95}Nb, and ^{99}Mo/^{99}Tc. Some reports from nuclear power plants and other dischargers, as well some research and monitoring reports, present the given combinations as single radionuclide entries. These sources report the total activity of the combination (a sum of the activities of the two radionuclides), but either do not or cannot disaggregate the data and assign the separate activities to each radionuclide in the pair. The second radionuclide in each pair is a decay product of the first, and the pair has assumed a special status in the radionuclide literature. However, most of those elements even when released to the water column do not enter biota. Therefore, only a small subset of the elements of the Table 25 are discussed individually.

Because of its importance to the Great Lakes, tritium is discussed separately in this section. For radionuclides other than tritium, the Task Force has prepared Table 27 bioaccumulation and biomagnification factors. These factors are based mainly on data for the stable isotopes of the elements, and where possible, from studies on the radioactive isotopes of the elements. All data were from studies of organisms found in lakes and especially the Great Lakes. Among the early available collection of freshwater data, Cowgill's work from 1973 to 1980 in Linsley Pond North Brantford, Connecticut, although not on the Great Lakes, provides one of the largest and most systematic studies of the uptake of chemical elements in plants and some animal species. Cowgill studied all detectable elements in the plants, animals, waters, and landscape materials of her study area (Cowgill 1970, 1973*a*, 1973*b*, 1974*a*, 1974*b*, 1976; Hutchinson 1975). She reported data as total elemental concentration (stable plus unstable) with no consideration as to isotopes. She compared her data with marine and oceanic studies because of a shortage of other data sets on elemental accumulations in plants and animals. Following her successful work on Linsley Pond, she turned her attention to similar studies in many aquatic systems ranging from laboratory cultures to entire marshlands in locations all over the world (Cowgill and Prance 1982; Cowgill et al 1986). The Task Force gratefully acknowledges her guidance in assessing the relationships among elements in aquatic species.

The Task Force also had access to a classic data set of Copeland et al (1973) for fish in the vicinity of nuclear power plants on Lake Michigan, and reporting on all of the elements that were quantifiable. Many reports cite this special compendium of Lake Michigan data, including the National Council on Radiation Protection (NCRP, #76 and #126), but often through secondary sources.

TABLE 25
RADIONUCLIDES OF IMPORTANCE FOR INVENTORIES IN BIOTIC COMPARTMENTS IN THE GREAT LAKES

Radionuclide	Half-Life	Category (I, Intermediate) (II, Long-lived)	Method of Production				
			Fission	Activation	Decay	Primordial	Cosmogenic
^{3}H	12 years	II	X	X			X
^{10}Be*	2.5 × 10^{6} years	II					X
^{14}C	5730 years	II		X			X
^{32}Si	650 years	I		X			X
^{32}P*	14.3 days	I		X			X
^{33}P*	25 days	I		X			X
^{40}K*	1.28 × 10^{6} years	II				X	
^{45}Ca	16.5 days	I		X			X
^{51}Cr*	27.8 days	I		X			
^{54}Mn*	303 days	I		X			
^{55}Fe*	2.6 years	I		X			
^{57}Co*	270 days	I		X			
^{58}Co*	71.3 days	I		X			
^{60}Co*	5.26 years	I		X			
^{63}Ni	92 years	II		X			
^{63}Zn	243 days	I		X			
^{86}Rb*	18.6 days	I		X			
^{87}Rb*	4.8 × 10^{10} years	II		X			
^{89}Sr*	52 days	I		X		X	
^{90}Sr*	28.1 years	II		X			
^{91}Y	58.8 days	I		X			

TABLE 25 (continued)
RADIONUCLIDES OF IMPORTANCE FOR INVENTORIES IN BIOTIC COMPARTMENTS IN THE GREAT LAKES

Radionuclide	Half-Life	Category (I, Intermediate) (II, Long-lived)	Method of Production				
			Fission	Activation	Decay	Primordial	Cosmogenic
^{95}Zr*	65 days	I	X				
^{95}Nb*	35.1 days	I	X				
^{95}Zr/^{95}Nb (combination)		I	X				
^{99}Tc*	2.1×10^5 years	I	X				
^{99}Mo/^{99}Tc (combination)		II	X				
^{103}Ru	39.6 days	I	X				
^{106}Ru	367 days	I	X				
^{110m}Ag	253 days	I	X				
^{113}Sn	115 days	I	X				
^{117m}Sn	14 days	I	X				
^{129}I*	1.7×10^7 years	II	X				
^{134}Cs*	2.05 years	I	X				
^{136}Cs*	13 days	I	X				
^{137}Cs*	30.23 years	I	X				
^{137}Ba	12.8 days	I	X				
^{140}Ba/^{140}La (combination)		I	X				
^{141}Ce*	33 days	I	X				
^{144}Ce*	285 days	I		X			
^{210}Pb	223 years	II			X	X	
^{226}Ra	1600 years	II			X	X	
^{232}Th	1.4×10^{10} years	II				X	
^{238}U	4.5×10^9 years	II				X	

Note: A radionuclide with an asterisk means that the element associated with this isotope is discussed in the bioaccumulation chapter.

Tritium

As previously noted, tritium is not routinely monitored in biological compartments. An early study, Rosenthal and Stewart (1971), and an important research project of the International Atomic Energy Agency (IAEA) with three studies, Blaylock and Frank (1979), Kirchmann et al. (1979), and Adams et al. (1979) provide the basic information.

Rosenthal and Stewart (1971) examined two algal species, an aquatic macrophyte, a *Daphnia*, and three species of freshwater snails in a small pond. Their work has two important characteristics: first, that the tritium which bound to biological tissues ("tissue bound tritium" or TBT) never exceeded 7% of the tritium taken up by the biota, and second, that the species studied occur in the Great Lakes Basin. The experiments ran a sufficiently long time relative to the life cycles of the biota to assure that the results could likely represent a steady-state bioaccumulation level. Their data also show that the levels of TBT may not show species selectivity between plants and animals, although plants seem to accumulate slightly more tritium than animals.

Rosenthal's work shows the need to understand the "tritium terminology." Tritium moves environmentally mainly as a tritiated water molecule, HTO. Discussions of biota tend to emphasize the different forms of HTO in organisms. Text Box 1 gives a short lexicon.

TEXT BOX 1
TRITIUM TERMINOLOGY

(1) Unbound HTO: molecularly intact and identifiable tritiated water which can move with a bulk water phase, and exhibits no surface, electrostatic, colloidal, hydrogen bonding, or other effects with another phase or a chemical component of its own phase other than bulk water. In some reports, it may be referred to as tritium in "free water."

(2) Bound HTO: molecularly intact and identifiable tritiated water which exhibits various kinds of observable surface attraction, hydrogen bonding, electrostatic bonding or other bonding effects associated with another phase or some chemical component other than bulk water in its own phase. This is sometimes referred to as "tritium in the free tissue water or TFTW."

(3) Combustion HTO: tritiated water which is chemically released upon combustion of biological material.

(4) Organically Bound HTO: tritium bound to biological molecules and cellular organic materials. This is sometimes referred to "tissue-bound tritium or TBT."

(5) Tritium Units: The ratio of the tritium content of a system to its stable hydrogen content as multiples of 10^{-18} which is the ratio of the natural occurrence of atoms of tritium to the atoms of stable hydrogen. These are sometimes abbreviated "TU."

Conversion factors: 1 TU = 10^{-18} atoms of ^{3}H/atoms stable H
1 TU implies a concentration of 0.12 Bq/L or 3.24pCi/L.

From the Text Box, the combustion HTO is a form of organically bound tritium. (This form of tritiated water appears to exert most of the adverse toxicological effects on biota.) Organisms can rapidly excrete unbound HTO and mobilize physiological systems to excrete less rapidly bound HTO. However, organisms retain organically bound HTO indefinitely and lose tritium by radioactive decay.

Blaylock and Frank (1979) examined some plants and animals from a pond on the Oak Ridge Reserve (Tennessee). Kirchmann et al. (1979) added several freshwater and marine species to those previously studied, including a salmonid, and some information on tritium apportionment in cellular compartments. Adams et al. (1979) reported on tritium uptake by Great Lakes aquatic biota in a freshwater marsh system in Lake Erie, near the Davis–Besse nuclear power plant. These studies used mutually agreed upon protocols and methods that assured that their data were statistically compatible (could be pooled statistically, because they come from the same "universe of data"); reported tritium bound to tissues on a dry weight basis, providing consistency and continuity with previous data of early investigators on bioaccumulation; and identified clearly the species studied taxonomically and tissues and substructures examined. Tables 26a–d present the data of Blaylock and Adams (1979), Rosenthal and Stewart (1971), Kirchmann et al. (1979), and Adams et al. (1979). The Task Force recalculated the bioaccumulation factors for Blaylock and Adams from the original paper. The Task Force only presents the data from North American species from Kirchmann's study.

Tritium levels in organisms track the tritium levels of the environment. Bioaccumulation factors of unity for tritium are conservative.

Tritium uptake in aquatic biota usually tracks the environmental levels. Some investigators note that tritium in organismal tissues quickly "equilibrate" with the tritium content of the surrounding water, but that view is simplistic. The data show that biota can excrete unbound tritium as HTO rather rapidly, and bound tritium as HTO more slowly, but may not be able to excrete tritium as organically bound HTO. Thus the view of "equilibration" is simplistic since tritium targets DNA and RNA, and through isotope exchange, tritium replaces a stable hydrogen on a nucleotide as organically bound HTO in the cell nucleus or extranuclear DNA in cytoplasm or organelles such as plasmids and chloroplast. "Equilibration" masks the isotope exchange on biomolecules because the exchange process occurs after uptake and at a much slower time scale.

From the available information, the Task Force affirms a bioaccumulation factor of unity (1) is conservative and confirms previous recommendations from marine studies.

The retention of 10% of the tissue-bound tritium level in organisms suggests a method of estimating an inventory of tissue-bound tritium in biota.

The previous information suggests a method to estimate a tritium inventory for aquatic organisms: as the product of the biomass and 10% of the environmental level of tritium. However, such an inventory calculation has a practical limitation in that most environmental tritium data for waters are reported at or below a level of detection, variably 100–200 pCi/L, depending on the instrumentation. Thus, the method only provides an upper bound estimate of the biological inventory of tritium.

TABLE 26a
TRITIUM ACCUMULATION IN AQUATIC ORGANISMS — PART I

Rosenthal and Stewart (1971)	Tritium Activities (pCi/mL or pCi/g)				Bioaccumulation Factors		
Plants	I: Lake Water	II: Unbound	III: Bound	IV: Food	V: R_1 (III)	VI: R_2 (III/I)	VII: R_3 (IV/III)
Chlamydomonus reinhardi	963	887	400	0.921	0.415		
Lemna minor	15.9	16.2	69	1.02	0.439		
Animals							
Daphnia galeata	892	928	482	511	1.4	0.540	0.9543
Lymnaea reflexa (snail)	774	46	845	511	1.09	1.09	1.65
Helisoma trivolus (snail)	15.1	4	2.7	511	0.26	0.53	0.0052

Adams et al. (1979)	Tritium Activities (pCi/mL or pCi/g)			Bioaccumulation Factors	
Plants	I: Lake Water	II: Unbound	III: Bound	IV: R_1	V: R_2
Cladophora sp.	609.7 ± 2.6	609.6 ± 6.6	78.9 ± 11.6	1.00	0.12 (0.38)
Spirodela polyrhiza	407.7 ± 8.3	413.0 ± 7.5	95.3 ± 14.1	0.98	0.23
Potamogeton foliosus	423.5 ± 9.9	422.9 ± 2.4	133.4 ± 20.4	0.99	0.31
Typha latifolia					
Stem	646.4 ± 8.3	361.1 ± 4.1	78.8 ± 7.6	0.56	0.12 (0.25)
Rhizome	646.4 ± 8.3	399.3 ± 2.5	75.4 ± 7.5	0.62	0.12

TABLE 26a (continued)
TRITIUM ACCUMULATION IN AQUATIC ORGANISMS — PART I

Adams et al. (1979)	Tritium Activities (pCi/mL or pCi/g)			Bioaccumulation Factors	
Animals	I: Lake Water	II: Unbound	III: Bound	IV: R_1 (II/I)	V: R_2 (III/I)
Plathemis lydia (dragonfly)	646.4 ± 8.3	527.1 ± 5.2	138.6 ± 39.0	0.82	0.21
Physa heterostropha (pond snail)	646.4 ± 8.3	555.7 ± 7.2	76.0 ± 29.6	0.86	0.11
Gambusia affinis (mosquitofish)	646.4 ± 8.3	555.3 ± 10.4	186.3 ± 14.3	0.86	0.29 (0.56)
Lepomis macrochirus (bluegill) muscle	403.2 ± 10.3	409.5 ± 6.7	187.9 ± 18.3	1.02	0.46 (0.69)
Micropterus salmoïdes (largemouth bass) muscle	403.2 ± 10.3	412.6 ± 7.0	166.2 ± 13.9	1.02	0.42(0.61)

Notes: (1) Locations: Rosenthal and Stewart — laboratory; Adams et al. — White Oak Lake at Oak Ridge, Tennessee (USA)

(2) Data from Rosenthal and Stewart are recalculated (original data in dpm/mL and dpm/g). All bound tritium data are on a dry weight basis.

(3) R_1 and R_2 are the bioaccumulation factors obtained by dividing the numbers in the indicated columns (Roman numerals separated by the slash). Column V entries in parentheses are as originally reported by Adams et al., which correct for specimen density.

(4) Data for *Gambusia affinis* are for the whole animal.

(5) Food chain relations from Adam's et al. data are undetermined. None of the listed animals species is known to consume any of the listed plant species. *Physa heterostropha*, epiphytic on macrophytes, eats algae. Algae of genus *Cladophora* are not usually desirable food for invertebrates and fishes. Dragonflies (*i.e.*, *Plathemis lydia*), consume smaller invertebrates rather than plants.

(6) References: adapted from Tables II and III (Blaylock and Frank 1979), with recalculation of bioaccumulation factors. See note 3.

TABLE 26b
TRITIUM ACCUMULATION IN AQUATIC ORGANISMS — PART II
GROWTH OF THE ALGA *Scenesdmus obliquus* ON LIQUID EFFLUENTS CONTAINING TRITIUM FROM NUCLEAR FACILITIES

	I	II	III
Effluent source	Initial tritium in culture medium (nCi/mL)	Tritium in water of dry tissue (μCi/mL)	Combustion *R* (II/I)
Radiochemical Lab	52	1.41	27.1
Radiochemical Lab	76.5	4.10	53.6
Radiochemical Lab	70.3	3.08	43.8
Nuclear Power Plant			
SEMO	5.1	4.60	0.90
SENA	16.6	14.5	0.87

Notes: (1) Location: Brussels, Belgium.

(2) Exposure time of algae: 12–42 days

(3) Distribution of tissue bound tritium (TBT) in organismal compartments: lipids, pigments and fatty acids, 58.3%; ether-soluble substances, ND (not detected); free amino acids and carbohydrates, 1.1%; protein hydrosylate, 34%; residue material, 6.6%. Nucleic acids contain more radioactively labeled hydrogens than lipids and lipid fractions. Ratios of activity in nucleic acid to lipid to lipid fraction, when referred back to the tritium content of the algal medium is 1.13:0.56:0.44.

(4) Nuclear effluents were filtered on Millipore (0.45 μm) before mixing with culture medium.

(5) The specific activity ratios for the effluents from two nuclear power plants (known as SEMO and SENA) suggest, according to the reference, an absence of biologically bound tritium in the filtered effluents. Radioactivity was reported from the filtered residue suggesting the uptake of tritium by microorganisms.

(6) Reference: adapted from Kirchmann et al (1979), especially Tables I and VI.

TABLE 26c
TRITIUM ACCUMULATION IN AQUATIC ORGANISMS — PART III
TRITIUM UPTAKE FOR FISHES GROWN IN LABORATORY AQUARIA OR IN OUTDOOR MESOCOSM POOLS

Species		Exposure Conditions		Specific Activity Ratio	
		Medium	Days in HTO	^{3}H in food	Commercial Food
Carassius auratus		Aquarium	32	0.13–0.25	
(goldfish)		Pool	173–214	0.15 [4]	0.27
Salmo gairdneri	Dead eggs[5]	Aquaria	1		0.39
(rainbow trout)	Dead fry	Aquaria	32		0.42
	Fish	Aquaria	140		0.15 [4]
Salmo trutta forma *fario*	Adult	Aquaria	5	0.16 [4]	0.15 [4]
(brown trout)	Adult	Aquaria	10		0.42
Gambusia affinis	Young	Aquaria	21	0.36	
(mosquitofish)	Young	Pool	11	0.56	0.37
	Young	Pool	39		
	Adult	Aquaria	60	0.52	
	Adult	Aquaria	203	0.63	
	Adult	Pool	58	0.88	
	Adult	Pool	93		0.51
Ictalurus lacustris	Dead eggs	Aquaria	3		0.41
(channel catfish)	Small fish	Aquaria	133		

Notes: (1) Locations: Berkeley, California; Las Vegas, Nevada; Brussels, Belgium.
(2) Exposure times: 10–30 days in Belgium, up to 203 days in the United States.
(3) Tritiated food in tests in Belgium; commercial food in tests in Belgium and the United States.
(4) Data reported for muscle tissue; data without footnote are for whole animal.
(5) Fertilized eggs for *Salmo gairdneri* placed in tritiated water when received.
(6) Specific activity ratio is the activity in dry tissue divided by that in the water medium.
(7) All tissue data are on a dry weight basis.
(8) Reference: adapted from Kirchmann et al (1979), Table III, notes and text.

TABLE 26d
TRITIUM ACCUMULATION IN AQUATIC ORGANISMS — PART IV
SPECIES STUDIED IN NAVARRE MARSH OF LAKE ERIE

I. Experimental conditions: 2-ha section of marsh. Earthen dike encloses 10,000 m^3 of water (depth of 50 cm; water surface area of approximately 20,000 m^2) . 10 Ci HTO added to marsh and the system followed for one year.

II. Plant species:

(a) Smartweed (*Polygonum lapathifolium*). Bound and unbound HTO follow the declining pattern of HTO in the marsh water. Evapotranspiration accounts mostly for tritium loss. Activity ratios of tritium in the tissue of the plant and marsh water are typically 0.3 or less throughout the observed period reported.

(b) Pickerelweed (*Pontederia cordata*). Follows the same pattern as smartweed, but activity ratios of bound HTO to marsh water tritium approach 1.0 at several times. Activity ratios for unbound HTO vary from 0.02 to 0.6.

(c) Pondweed (*Potamogeton crispus*). Follows a similar pattern to smartweed. Activity ratios over a long period of observation for bound and unbound HTO both range from 0.6 to 1.0, but then bound HTO ratios decline to near zero near the end of the observation period while the unbound HTO activity ratios remain in a range of 0.3–0.6.

III. Animal species:

(a) The crayfish (*Procambarus blandingi*) accumulates maximum activity in muscle and viscera within 2 and 3 days directly, then the both bound HTO and unbound HTO decline rapidly. Maximum activity ratios of Bound HTO/Unbound HTO: muscle, 0.34; viscera, 0.23. The unbound HTO follows the marsh water, with activity ratios of close to 1. The bound HTO never reaches an activity level relative to marsh water of greater than 0.2, declines to less than 0.1 within 2 months, and hovers near zero for the rest of the observation period.

(b) Carp (*Cyprinus carpio*) accumulates maximum activity in muscle and viscera within 4 hours of exposure then declines. Maximum activity ratios Bound HTO/Unbound HTO: muscle, 0.25; viscera, 0.39. The unbound HTO follows the same pattern as crayfish, with activity ratios close to 1 during much of the observation period and through its end. The bound HTO activity ratio never exceeds 0.1, remains relatively constant through much of the observation period, then declines to zero near the end of that period.

(c) Bluegill (*Lepomis macrochirus*) accumulates maximum activity in muscle and viscera within 1 day. Maximum activity ratios Bound HTO/Unbound HTO: muscle, 0.35; viscera, 0.38. The unbound HTO tracks the marsh water activity decline for the rest of the period (activity ratios of about 0.8), while the bound HTO declines rapidly to near zero within 70 days for both muscle and viscera.

IV. To track the tritium in the marsh water, the following data are extracted from the paper by Adams et al:

Days Post-Treatment	Tritium concentration in marsh water (dpm/L)	Days Post-Treatment	Tritium concentration in marsh water (dpm/L)
0	25,000	40	13,000
2	25,000	70	11,000
4	23,000	100	8000
9	20,000	170	5000
		220	>2000

Notes: (1) Reference: adapted from Adams et al. (1979), especially Figures 1–4 and text.
(2) Tritium count data are rounded out to the nearest thousand.
(3) No estimate of biomagnification factors from the species studied because of a lack of an established food-chain relationship.
(4) dpm, disintegrations per minute.

TABLE 27
BIOACCUMULATION FACTORS AND BIOMAGNIFICATION FACTORS FOR VARIOUS ELEMENTS IN AQUATIC BIOTA

	Bioaccumulation				Biomagnification		
Elements	Plants/ Water	Invertebrates/ Water	Fishes/ Water	Plants/ Sediments	Plants/ Invertebrates	Fishes/ Plants	Fishes/ Invertebrates
Beryllium	3000	—	—	—	0.05	—	—
Phosphorus	NA	NA	NA	NA	NA	NA	NA
Potassium	1000–3000	—	800	—	1–2	—	—
Chromium	50,000	50,000	—	—	1	—	—
Manganese	50,000	—	—	—	0.5	—	—
Iron	50,000	5000	—	—	0.1	—	—
Cobalt	300,000	60,000	—	—	1	—	—
Rubidium	10,000	12,000	—	—	1	—	—
Strontium	1000	—	—	—	1.5	—	—
Zirconium	2000	2000	—	—	1.5	—	—
Niobium	20,000	20,000	—	—	1.5	—	—
Iodine	2000	1000	—	—	0.4	—	—
Cesium	20,000	10,000	50	—	0.5	—	—
Cerium	20,000	—	—	—	1	—	—

Notes: NA, not available.
(—) The Task Force was unable to find data.

Univalent Elements

The univalent elements sodium, potassium, lithium, rubidium, cesium, silver, chlorine, bromine, and iodine are found in freshwater lakes and their biota. The Task Force was especially interested in potassium, rubidium, cesium, and iodine.

The isotope ^{40}K is a major natural source of radioactivity. It accounts for as much as 40% of the radioactivity present in biological tissues (humans included). Potassium is a macro element with important physiological functions, and therefore, its presence exerts an important control function on the cycling of other elements. Measurements of ^{40}K in Great Lakes biota appear in several documents including the annual reports of various nuclear power plants. Since geological data do not clearly show a selective or differential enrichment of potassium isotopes in geological repositories (*i.e.*, glaciers, mineralized soils, and volcanic rock), a simple inventory estimate for ^{40}K depends on the assumption that the ratio of radioactive potassium to stable potassium in biological compartments and substrates is numerically the same as the ratio of radioactive potassium to stable potassium in geological strata.

Since freshwaters are impoverished in sodium (by definition), isotopes of sodium although limnologically important are not discussed. The naturally occurring radioactive isotopes of sodium have not been reported on in biota. An inventory procedure similar for the one ^{40}K does not apply to radioactive isotopes of sodium.

Rubidium and Cesium

Rubidium accumulates in plants often with the ability to substitute for potassium and sodium in selected chemical matrices. Isotopes of rubidium are emitted directly by nuclear power plants to the Great Lakes as well as forming as decay products of noble gas radionuclides

Although cesium chemistry parallels sodium and potassium chemistries, cesium does not easily replace either sodium or potassium in various chemical matrices because its atomic and ionic radii are larger than those of sodium and potassium. The element's rarity in nature also limits its biological availability.

The most extensive data base for bioaccumulation of an artificial radionuclide exists for ^{137}Cs. This long-lived isotope (half-life: 30 years) was the first long-lived isotope in nuclear fallout studied in biota as part of biological monitoring and the study of the effect of radio isotopes on biota. The important compilations on radiocesium uptake are the studies of Blaylock (1982), Joshi (1984), and Elliott et al. (1984). The latter two references discuss Great Lakes fishes. Most of the compilations from these authors were from research components of monitoring studies. Further, dischargers of radionuclide materials in the Great Lakes Basin must monitor for ^{137}Cs in biological substrates, although the data have deficiencies that limit its use for biocompartment inventory calculations.

The Task Force has also examined monitoring data from nuclear power plants for ^{137}Cs in fishes. Tables 28 and 29 present data for the bioaccumulation factors for ^{137}Cs for Lakes Huron, Erie, and Ontario. All of the bioaccumulation factors in Table 29 have the same order of magnitude, although fishes in Lake Huron have higher bioaccumulation factors than fishes in Lake Ontario. These factors should **not** be assumed to hold for the two Lakes over all periods of time. Joshi (1984) calculated bioaccumulation factors from several fish studies going back to 1976. The data in Table 29 show that the ratio of ^{137}Cs to ^{40}K is about 0.01. This suggests a way to calculate an inventory for ^{137}Cs. From the estimate of the inventory for ^{40}K, estimate the inventory for ^{137}Cs by adjusting the estimate for ^{40}K by 0.01.

Given the small bioaccumulation rates for radionuclides of cesium, the final repository for radiocesium from fallout and liquid and gaseous discharges to the Great Lakes is sediments. Although cesium salts are relatively soluble in aquatic media, the attachment of the cesium compounds to other particulates provides a mechanism to reach sediments with minimal physicochemical interaction with aquatic media.

Silver

One radionuclide of silver, ^{110m}Ag (half-life: 233 days), consistently appears in the nuclear discharges to the Great Lakes. The radionuclide has been reported every year in the gaseous and liquid discharges of at least eight nuclear power plants in the Great Lakes over the period of 1980–1993. It persists long enough for silver to cycle through Great Lakes biota following its discharge. However, there is no procedure derivable from the data with which the Task Force could estimate and inventory of this isotope in biological compartments.

Fluorine, chlorine, and bromine

There are no radionuclides of fluorine that need to be discussed. Radionuclides of chlorine are mainly cosmological, although ^{36}Cl sometimes appears as a fission product. The radionuclides of bromine are fission products and are associated with fuel rods and would not be expected to be discharged routinely to the Great Lakes.

Iodine

Freshwater and marine biota both accumulate iodine. The environmental levels of iodine are greatest in the oceans, and marine plants, invertebrates and vertebrates accumulate it to levels greater than freshwater or terrestrial organisms. The low levels of iodine in freshwater limit its uptake almost uniquely to plants and vertebrates, the latter biota having thyroid glands. There are very limited data on the uptake of iodine by freshwater invertebrates. Bioaccumulation factors for iodine of about 500 for plants and biomagnification factors of about 0.05 for zooplankton and insects seem appropriate.

TABLE 28
CESIUM ACCUMULATION IN GREAT LAKES BIOTA – PART I
^{137}Cs IN GREAT LAKES FISHES
(adapted from International Joint Commission and other agency reports)

Year	Species	Location	Average ^{137}Cs Activity (Bq/kg)	Bioaccumulation Factor
Lake Huron				
1981	Walleye	Blind River	9.78 ± 0.42 (6)	
1981	Sturgeon	Blind River	3.36 ± 0.25 (4)	
1982	Lake trout	North Channel	8.22 ± 0.17 (3)	
Lake Erie				
1982	Walleye	Western Basin	0.862 ± 0.085 (3)	1556
Lake Ontario				
1976	Rainbow trout	Ganaraska River	2.4	3528
1977	Rainbow trout	Ganaraska River	2.0	2391
1978	Rainbow trout	Ganaraska River	2.2	2354
1980	Rainbow trout	Ganaraska River	2.7	
1981	Rainbow trout	Ganaraska River	1.40 ± 0.13 (9)	1700
1982	Rainbow trout	Coburg	1.5 ± 0.1	1414
1982	Lake trout	Coburg	1.5 ± 0.1 (3)	1425
1982	Lake trout	Niagara on the Lake	1.60 ± 0.12 (4)	1490
1982	Lake trout	Oswego	1.60 ± 0.4 (2)	

Notes: (1) References: International Joint Commission (1983, 1987*b*); Joshi (1984, 1985, 1986, 1987, 1988*a*, 1988*b*).

(2) Numbers in parentheses are number of fishes used in the averaging. If no number appears in parentheses, then either only one measurement was reported, or the source of the data did not qualify the information in some manner.

(3) Data are from Environment Canada and New York State Department of Health.

(4) Radioactive measurements made on wet weight basis.

(5) Bioaccumulation factors from Joshi (1984). Bioaccumulation factors relate to open water levels or ambient levels at collection site, depending on the available data.

TABLE 29
CESIUM ACCUMULATION IN GREAT LAKES BIOTA — PART II ^{137}Cs IN GREAT LAKES FISHES; STUDIES AT HWR NUCLEAR GENERATING STATIONS
(adapted from operating reports of Ontario Hydro)

Year	Lake and Station	Sample or Substrate	Activity (Bq/L or Bq/kg)		^{137}Cs/^{40}K	Sample or Substrate	Bioaccumulation Factors	
			^{137}Cs	^{40}K			^{137}Cs	^{40}K
1992	Ontario	Trout	1.07 ± 0.15	120 ± 4	0.0089	Trout/water	*	857
	Darlington	Whitefish	0.56 ± 0.15	119 ± 4	0.0047	Whitefish/water	*	850
	(Provincial Park)	Sucker	0.18 ± 0.15	105 ± 6	0.0017	Sucker/water	*	750
		Water	<0.003	0.14 ± 0.03		Water/sediments		0.000389
		Sediments	2.5 ± 1.3	360 ± 30	0.0069			
1992	Ontario	Trout	0.96 ± 0.18	124 ± 6	0.0077	Trout/water	*	886
	Darlington	Whitefish	0.48 ± 0.18	126 ± 6	0.0038	Whitefish/water	*	900
		Sucker	0.23 ± 0.11	109 ± 4	0.0021	Sucker/water	*	779
		Water	<0.003	0.14 ± 0.04		Water/sediments		0.000342
		Sediments	<1.2	410 ± 30				
1992	Ontario	Trout	1.04 ± 0.22	137 ± 7	0.0076	Trout/water	69.3	979
	Pickering	Whitefish	0.63 ± 0.15	120 ± 4	0.0053	Whitefish/water	42.0	857
	("A" Discharge)	Sucker	1.15 ± 0.22	121 ± 7	0.0095	Sucker/water	76.7	864
		Water	0.015 ± 0.004	0.14 ± 0.04	0.107	Water/sediment	0.00091	0.000342
		Sediments	16.5 ± 1.5	410 ± 30	0.0402			
1992	Ontario	Trout	0.92 ± 0.18	132 ± 4	0.0069	Trout/water	*	776
	Pickering	Whitefish	0.85 ± 0.22	136 ± 6	0.0063	Whitefish/water	*	800
	("B" Discharge)	Sucker	1.00 ± 0.22	131 ± 6	0.0076	Sucker/water	*	771
		Water	<0.003	0.17 ± 0.05		Water/sediment		0.000395
		Sediments	7.0 ± 1.3	430 ± 30	0.0162			

TABLE 29 (continued)
CESIUM ACCUMULATION IN GREAT LAKES BIOTA — PART II:
^{137}Cs IN GREAT LAKES FISHES; STUDIES AT HWR NUCLEAR GENERATING STATIONS
(adapted from operating reports of Ontario Hydro)

Year	Lake and Station	Sample or Substrate	Activity (Bq/L or Bq/kg) ^{137}Cs	^{40}K	^{137}Cs/^{40}K	Sample or Substrate	Bioaccumulation Factors ^{137}Cs	^{40}K
1992	Ontario	Trout	1.00 ± 0.15	104 ± 3	0.0096	Trout/water	0	612
	Pickering	Whitefish	0.74 ± 0.22	133 ± 6	0.0056	Whitefish/water	*	783
	(Duffins Creek)	Sucker	0.96 ± 0.18	153 ± 4	0.0063	Sucker/water	0	900
		Water	<0.003	0.17 ± 0.04				
		Sediments	8.5 ± 1.4	390 ± 20	0.022	Water/sediment		0.00044
1992	Ontario							
	Pickering	Rainbow trout	<0.15	128 ± 4 (w)				
	(Coolwater Farms)	Rainbow trout	0.2 ± 0.1	117 ± 4 (s)	0.0017			

Limiting Values of Bioaccumulation Factors for ^{137}Cs in fishes relative to water based on LLD of the radionuclide of 0.003 Bq/L for Lake Ontario Nuclear Generating Stations

Trout/water	357 (Darlington: Prov. Park)
Trout/water	320 (Darlington: NGS)
Trout/water	307 (Pickering: B)
Trout/water	333 (Pickering: Duffins Creek)

Average geochemical ratios ^{137}Cs/^{40}K factors for species monitored at HWR facilities

Lake and Station	Species	^{137}Cs/^{40}K	Species	^{137}Cs/^{40}K	Species	^{137}Cs/^{40}K	Species	^{137}Cs/^{40}K
Lake Ontario:								
Darlington	Trout	0.0083	Whitefish	0.0043	Sucker	0.0019		
Pickering	Trout	0.0083	Whitefish	0.0053	Sucker	0.0078		
Average for Lake Ontario fish:	Trout	0.0083	Whitefish	0.0048	Sucker	0.0048	All fishes	0.0060

Average Bioaccumulation factors for species monitored at HWR facilities on Lake Ontario

	^{137}Cs	^{40}K		^{137}Cs	^{40}K
Darlington			Pickering		
Trout/water	*	872	Trout/water	69	789
Whitefish/water	*	875	Whitefish/water	42	813
Sucker/water	*	765	Sucker/water	77	845
Average all fish/water	*	837	Average all fish/water	63	816

TABLE 29 (continued)
CESIUM ACCUMULATION IN GREAT LAKES BIOTA — PART II:
^{137}Cs IN GREAT LAKES FISHES; STUDIES AT HWR NUCLEAR GENERATING STATIONS
(adapted from operating reports of Ontario Hydro)

Average geochemical ratios for ^{137}Cs/^{40}K for species monitored at HWR facilities on Lake Huron

Station	Species	^{137}Cs/^{40}K	Species	^{137}Cs/^{40}K	Species	^{137}Cs/^{40}K	Species	^{137}Cs/^{40}K
Bruce	Trout	0.0185	Walleye	0.0238	Bass	0.021	All fish	0.0194

Average Bioaccumulation factors for species monitored at HWR facilities on Lake Huron

Station	Species	^{137}Cs	^{40}K
Bruce	Walleye/water	*	1063
	Bass/water	*	1123
	Trout/water	*	1318
	Average all fish/water	*	1168

Comparisons of Lakes Ontario and Huron

Average geochemical ratios for ^{137}Cs/^{40}K Lake Ontario/Lake Huron				Average bioaccumulation factors ^{137}Cs and ^{40}K Lake Ontario/Lake Huron		
Trout	0.45	All fish	0.31	All fish/water	*	1.41

Notes: (1) Sediment data are dry weight; fish data are wet weight

(2) Symbols: (w) samples taken in winter–spring period (January to June); (s) samples taken in summer–autumn period (July to December); (*) cannot calculate the number from information given.

(3) Averages of bioaccumulation factors are calculated when there are two or more results for a given species of fish. The overall average bioaccumulation factor for fishes in a lake averages all data from that lake without regard to the number of entries per species.

(4) Bioaccumulation factors reported for ^{137}Cs based on one data set only.

(5) Ratio of bioaccumulation factors for ^{40}K for Lake Ontario and Lake Huron comparison obtained by taking the ratio of the averages of all of the bioaccumulation factors for the fishes of each lake.

Divalent Elements

The divalent elements beryllium, magnesium, calcium, strontium, and barium are all found in the waters, sediment, and biota of the Great Lakes. The studies of two investigators, Cowgill (1973*a*, 1973*b*, 1974*a*, 1974*b*) and Yan et al. (1989) are most important.

Beryllium

Cowgill's data are the most comprehensive for beryllium. Because beryllium isotopes produced cosmogenically remain in the atmosphere for a long time, the appearance of radioactive beryllium in Great Lakes biota suggests that the beryllium probably originates from artificial sources, mainly reactions in a nuclear reactor. ^{7}Be has been occasionally documented in gaseous and liquid effluents of nuclear reactors, but as a very minor radionuclide.

Calcium and Magnesium

The radionuclides of calcium and magnesium are too short lived to consider biological inventories. However, the Task Force notes that chemical data for both elements are needed in discussing the behaviour of the radionuclides of other elements cycling within the Great Lakes, especially radionuclides of divalent elements in the same or adjacent families of the Periodic Table: beryllium and barium. Calcium also correlates with many other elements: titanium, phosphorus, iron, cerium, lanthanum, and most other rare earth elements. The correlation suggests that it exerts a control function on the cycling of many other elements in freshwater biota.

Strontium

Strontium has several important radionuclides. The element accumulates in plants, can biomagnify up the food chain, and thus presents a health hazard to humans through intake of food. Isotopes of strontium can be found in the water column because many strontium compounds are water soluble. Although strontium has bone as its target organ in vertebrates, the Task Force could not confirm that calcium has a control influence on either strontium uptake or biological cycling.

Barium

^{140}Ba is a fission product of some importance. It accumulates in biota, but the available data on barium levels in biota do not permit a separate calculation of the radioactive barium content. The Task Force did find that levels of barium in biota are correlated with levels of calcium and that high environmental levels of calcium and high tissue levels of calcium both block uptake and accumulation of tissue levels of barium.

Transition Elements

Zinc, manganese, iron, technetium, ruthenium, chromium, nickel, cobalt, and molybdenum belong to different chemical groups, but it is often easier to address them together. Their radionuclides are mainly activation products, but they are also produced by fission. Because their nuclear reactions involve interconversions, their methods of analysis usually provide data on all them virtually simultaneously. All of these elements accumulate in biota, but the pathways for technetium are unknown. That element has not been studied in aquatic biota despite the fact that its major ion, pertechnate $(TcO_4)^{1-}$ is water soluble and allows potential direct uptake by biota from the water column.

Zinc

The important radionuclide of zinc is ^{65}Zn (half-life: 244 days), which can cycle within the Great Lakes. Zinc is an essential trace element in nutrition, assuring its bioaccumulation in all species. From the available date on zinc levels in Great Lakes biota, the Task Force could not calculate a separate inventory for this radioisotope.

Chromium

The important radionuclide of chromium is ^{51}Cr. Chromium cycles through biological compartments in several valence states, two of which are very important in aquatic systems. The hexavalent (+6) state is water soluble and very toxic to most organisms. The trivalent (+3) state has a low water solubility and behaves as a trace micronutrient in certain tissue and organismal systems (Mertz 1967; see also other papers by Walter Mertz). Most environmental studies report a total chromium level without specification of the valence states. From the available date on chromium levels in Great Lakes biota, the Task Force could not calculate a separate inventory for this radioisotope.

Molybdenum

Molybdenum is a trace element nutrient needed by plants. It occurs in several enzymes associated with nitrogen fixation and the utilization of iron and sulfur in cellular metabolism. Two radionuclides, ^{95}Mo and ^{99}Mo, form as fission and activation products. Although ^{99}Mo has a half -life of only 6 hours, it decays to ^{99}Tc (half-life: 212,000 years) and thus plays an important role in estimating inventories for technetium. ^{99}Mo and ^{99}Tc are sometimes treated as a combined pair. From the available date on molybdenum levels in Great Lakes biota, the Task Force could not calculate a separate inventory for this radioisotope.

Iron and Manganese

The important radionuclides are ^{55}Fe (half-life: 2.6 years), ^{59}Fe (half-life: 45 days), ^{54}Mn (half-life: 303 days), and ^{56}Mn (half-life: 2.6 hours). The two radionuclides of iron and lower atomic weight radionuclide of manganese last long enough to cycle through Great Lakes ecosystems.

Depending on the pH of the freshwater system or the "local pH" (the acidity of surface materials) and the level of oxygenation of the system (aerobic versus anaerobic environment), iron and manganese hydroxides can form precipitates on the external surfaces of biological materials. Many analyses of plants that report very high iron and manganese levels may actually have reported crystalline ferric and manganese oxides as surface contaminants. Despite these chemical artifacts, both elements have major metabolic roles in organisms and approach a status of macronutrient rather than simple trace nutritional requirement. From the available date on iron and manganese levels in Great Lakes biota, the Task Force could not calculate separate inventories for the listed radioisotopes.

Ruthenium

Without nuclear technology, ruthenium would be of little interest to the Task Force. Two radionuclides are produced by fission and appear mainly in nuclear fuel processing operations: ^{103}Ru (half-life: 41 days) and ^{106}Ru (half-life: 1 year). The radionuclides can appear in both gaseous and liquid effluents. They were detected in the atmospheric fallout to the Great Lakes after the accident at Chernobyl, but not from the liquid discharges of radionuclides from nuclear power operations. Both radionuclides persist long enough for possible cycling within the biota of the Great Lakes, but the element has rarely been detected analytically in freshwater biota and reported in the literature. It has no known biological role, and despite its persistence, the available studies suggest that it moves mostly to sediments. The Task Force has not estimated a biocompartment inventory for ruthenium given its lack of observation in freshwater biota.

Cobalt

All radionuclides of cobalt are activation products. Four radionuclides, ^{57}Co, ^{58}Co, ^{59}Co, and ^{60}Co, are discharged to the Great Lakes, but two of them, ^{60}Co (half-life: 5.26 years) and ^{57}Co (half-life:270 days), last long enough to cycle biologically. The former has commercial use, and the latter is used in research.

Cobalt is an essential micronutrient that activates vitamin B_{12}. Therefore, cobalt uptake occurs in all aquatic biota. Some algae, macrophytes, and invertebrates can substitute cobalt for zinc in essential enzyme systems (Price and Morel 1990; see also other papers of François Morel), suggesting that Zn:Co geochemical ratios may be important in certain water bodies. From the available data on cobalt levels in Great Lakes biota, the Task Force could not calculate a separate inventories for cobalt radioisotopes.

Sulfur, selenium, and tellurium

Except for polonium, a decay product of transuranic elements, none of the Group VIb elements require inventories. The Task Force has previously discussed the cosmogenically produced radionuclides of sulfur. All radionuclides of selenium produced by nuclear activities in the Great Lakes Region except for ^{79}Se are very short lived, but the residuals of ^{79}Se are important only in considerations of the high-level waste inventories for fuel elements. The Task Force has no documentation that selenium radionuclides are released by nuclear facilities to the Great Lakes Basin. Despite the toxicity of tellurium, biota only show limited accumulation. Nor are radionuclides of tellurium documented in the releases from nuclear power plants, although such radionuclides would require consideration in the high-level waste inventories for fuel elements. Several isotopes of tellurium form as fission products. The volatility of many tellurium compounds is the factor explaining their appearance in the gaseous radionuclide emissions of fuel reprocessing operations.

Trivalent Elements

The trivalent elements include boron, aluminum, gallium, indium, and thallium. Boron and aluminium bioaccumulate in organisms, but there is no indication that they are discharged to the Great Lakes. Their congener elements of gallium, indium, and thallium do not have radionuclides that are produced in nuclear systems that are likely to be discharged to the Great Lakes.

Scandium, yttrium, lanthanum, and rare earths

The Group IIIa elements of the Periodic Table are rather unusual. All are rare in nature. Scandium often shows up in particulate matter sampled in the upper atmosphere, and has two radionuclides which form cosmogenically. But while scandium can bioaccumulate, there is no indication that its radionuclides are related to either natural background levels or radioactive discharges to the Great Lakes.

Yttrium, lanthanum, and the other rare earth elements, are well represented among the radionuclides formed in the nuclear fuel cycle. Many of these elements have primordial radionuclides, and almost all of the isotopes of rare earth elements are mildly radioactive. The behaviour of these elements in biota is not well understood. Cowgill's studies suggested that organisms may exert considerable selectivity on which elements they accumulate: besides cerium and lanthanum, and occasionally europium, the freshwater plants seem to favour the elements of even atomic number. They also accumulate in biota greatly compared with source materials, which often have levels undetectable by present methods.

Yttrium: Radionuclides of yttrium are fission products and the decay products of radioactive strontium. Two radionuclides of yttrium occur in discharges to the Great Lakes, ^{90}Y and ^{91}Y. The former has a very short half life, but the latter has a sufficiently long half-life to be of interest. Yttrium can accumulate in organisms, but the available data base is sparse; Cowgill's data are the most comprehensive. Yttrium's environmental cycling appears to follow its congener elements.

Lanthanum and Cerium: Lanthanum, although very toxic to aquatic biota, is detected in small amounts in aquatic biota along with other rare earth elements. Lanthanum phosphate, the compound expected in most fresh waters, is very insoluble, limiting its bioavailability. The decay of^{140}Ba to ^{140}La makes lanthanum of biological interest.

Cerium is the only other rare earth element besides lanthanum that is usually detected in biota. Cowgill's data probably form the most complete set on the stable forms of the element in freshwater biota. Cerium isotopes except ^{138}Ce, which is cosmogenic, are fission products and have been detected in Great Lakes waters. The two major radionuclides, ^{141}Ce and ^{144}Ce, usually appear in the aerosol content of gaseous emissions from nuclear power plants and occasionally in the liquid emissions. In nuclear fuel reprocessing operations, these radionuclides can appear equally likely in both gaseous and liquid emissions. Both isotopes were detected in the Great Lakes waters following the incident at Chernobyl.

Calcium appears to control the biological uptake of lanthanum and cerium as well as other lanthanides. The mechanism is unclear. It may block uptake directly by a mechanism similar to the one for barium or indirectly because of the relationship with phosphorus. Where uptake occurs, the Task Force has no indication that radionuclides of other rare earth elements behave differently with respect to calcium, and thus further discussion of individual lanthanides does not appear warranted.

The Task Force derived bioaccumulation factors of 20,000 for both lanthanum and cerium in freshwater plants; biomagnification factors were 2.4 in zooplankton and insects for lanthanum and 1.0 in zooplankton and insects for cerium.

Quadrivalent Elements

The quadrivalent elements are carbon, silicon, germanium, tin, and lead. Despite their common chemical grouping, their chemistry differs very markedly from element to element.

Carbon has already been discussed as ^{14}C. Silicon bioaccumulates in some species as an essential element in the skeletal structure of diatoms and foraminifera, usually in the amorphous mineralogical form of opaline phytoliths. There is no indication that radioisotopes of silicon are emitted to the Great Lakes. The two cosmogenically produced radionuclides of silicon, ^{31}Si and ^{32}Si, have unknown impact on the Great Lakes. Germanium isotopes can result from fission, but the discharge of germanium to the Great Lakes is largely undocumented.

Radionuclides of lead result from the decay of transuranics. The long life of ^{210}Pb and the concern about lead as a toxicant and air pollutant have led researchers to study this element and this particular radionuclide intensively. Thus, the information bases on radionuclide and stable lead are rather large, but the biological data on the radionuclide form of lead is not very extensive.

Tin

Tin is the most difficult quadrivalent element to consider. Very few data exist on its occurrence in aquatic systems and even less in biological compartments, Cowgill's data being the most comprehensive. Analytical methods to quantify tin in environmental media and substrates call for considerable skill and instrumental sophistication. The important radionuclides are ^{113}Sn (half-life: 115 days) and ^{117m}Sn (half-life: 14 days), and ^{126}Sn (half-life: 100,000 years). The first two radionuclides originate by fission and activation processes, the third by activation processes only. The first two have been occasionally documented in the Great Lakes, while third one has not, although its appearance would signal trouble. The activation sources are the zircalloy (a zirconium–tin alloy) cladding for nuclear fuel elements, making them important radionuclides from the nuclear fuel cycle. From the available data on tin levels in Great Lakes biota, the Task Force could not calculate a separate inventories for tin radioisotopes.

Quinquevalent Elements

The quinquevalent elements include arsenic, phosphorus, antimony, and bismuth. Transition quinquevalent elements are vanadium, niobium, and tantalum. Quadrivalent transition elements, specifically titanium, zirconium, and hafnium have biological characteristics that follow the quinquevalent elements. Data for biological uptake of antimony, bismuth, and tantalum are very rare or non-existent. Titanium and hafnium accumulate in plants, but there is no indication that their radionuclides are discharged to the Great Lakes. Titanium data assist in establishing inventories for many other elements including phosphorus, calcium, and the lanthanides.

Phosphorus

Of all of the elements of the Periodic Table, phosphorus holds a special place in the limnology of the Great Lakes. Starting in 1972, the removal of phosphorus from point sources and its quantification and management in non-point sources and sediments has guided programs to control eutrophication (the proliferation of aquatic plants which are stimulated by excess or luxuriant levels of nutrients, mainly phosphorus) of the Great Lakes. The chemistry and biology of phosphorus in the Great Lakes is rather complex subject to understand, and the control of eutrophication in the Great Lakes, while highly successful, has not achieved results sufficient to reduce the current control programs.

Phosphorus has two radioisotopes, both of which have cosmogenic as well as other sources. Although the isotopes have half-lives of the order of a few days to a few weeks, the chemical dynamics of phosphorus make all sources of phosphorus a special concern. Further, the early work on understanding the dynamics of phosphorus chemistry and biology in lakes used both of these isotopes as tracers.

In the section on sources of radioactivity, the data presented on secondary sources (*e.g.*, hospitals, university laboratories and reactors, commercial uses) indicated widespread use and discharge of both radioisotopes of phosphorus. For these reasons, phosphorus has received special attention in the Inventory of Radionuclides.

Zirconium and niobium

Zirconium and niobium are associated with the cladding material of fuel elements in nuclear power plants. Their radionuclides are both neutron activation and fission products. Radionuclides of zirconium, mainly ^{95}Zr, occurred in the fallout from atmospheric testing of nuclear weapons prior to 1963. Radionuclides of zirconium decay to radionuclides of niobium, and the combination ^{95}Zr/^{95}Nb sometimes appears as a single radionuclide in the emission reports of nuclear facilities. Both ^{95}Zr (half-life: 65 days) and ^{95}Nb (half-life: 35 days) are of potential environmental concern. The radionuclide, ^{94}Nb (half-life: 20,000 years), is one of the longest lived, but there are no indications that it is either discharged directly to the Great Lakes region or forms as a decay product of another radionuclide discharged to the Great Lakes. It has a very small fission yield and is included among the radionuclide inventories for spent nuclear fuel.

Both elements bioaccumulate. Cowgill's data are the most extensive and suggest niobium accumulates in plants more than zirconium. From the available data on zirconium and niobium levels in Great Lakes biota, the Task Force could not calculate separate inventories for their radioisotopes.

Vanadium

Vanadium accumulates. It is an essential micronutrient for certain plants and fungi. ^{50}V is a primordial radionuclide, and its inventory in biological tissue can be estimated by a similar procedure used for ^{40}K. Other radionuclides of vanadium form as both fission and activation products. Recent interest in vanadium by a number of researchers has provided a data base on vanadium uptake in plants and animals that offers possibilities in producing inventories for the biological compartments. However, there is only limited indication that vanadium radionuclides are released to the Great Lakes, and the only inventory that can be calculated for a radioactive isotope of vanadium is for its primordial radionuclide ^{50}V by multiplying the vanadium level in biota by 0.0025.

4 CONCLUSIONS

4.1 Adequacy of Monitoring

The Task Force concludes that:

(1) Monitoring meets the needs of the relevant atomic energy acts in the United States and Canada but is not designed to look at environmental cycling of radionuclides.

(2) Quality Assurance Protocols are also designed for compliance monitoring. Therefore, it is not possible to tell if nuclear plant monitoring is satisfactory to assure meeting the goals and objectives of the Great Lakes Water Quality Agreement.

Discussions

The information base used to assemble the inventories, notably the emissions data from nuclear facilities, and the monitoring data off-site of the facility but keyed to activities of the facility, has many problems. The Task Force reviewed the actual monitoring protocols (*i.e.*, directives, instrumentation, sampling plans, chemical analysis techniques, station and monitoring site locations, quality assurance considerations, data reporting and statistical analysis procedures), and found the following:

(A) All monitoring has as its primary goal to show that a given nuclear facility complies with the health, safety, and environmental requirements of its facility license. In turn, the health, safety, and environmental requirements in the license are dictated by the atomic energy legislation of each country. Thus, the Task Force concluded that the current state of monitoring is that of compliance.

(B) The atomic energy legislation of each country prescribes a maximum annual allowable human exposure to radiation as the basis for setting the environmental monitoring requirements for each individual radionuclide. The use of dose assessment models translate this exposure criterion into allowable discharges of specific radionuclides and types of energy.

(C) The dose assessment models used to derive the allowable discharges have a very limited relationship to the cycling of radionuclides for development of an inventory. The models make assumptions about the distribution of the activity of a given radionuclide in different environmental compartments and the fraction of that radionuclide's activity which is taken up by biota and assimilated and retained as opposed to taken up and then released, excreted, or otherwise removed. The models also make specific assumptions about the transfer of radioactivity from retained radionuclides in other biological compartments and the movements of the radionuclides through various foodwebs. This includes direct uptake by humans through drinking water or through intermediate uptake and bioaccumulation through food species.

(D) When monitoring environmental media, it is a particular characteristic of radionuclide measurements that the lower limit of detection for a given sample will depend on the amount of time lapsed between collection and analysis. This arises because the radioactivity in the sample continues to decay after sample collection, and all measured activities must be corrected back to the time of collection. Thus, the reported lower limits of detection may vary considerably from one laboratory to another, or even for measurements carried out in the same laboratory at different times after collection. For this reason it is not practical to use reported lower limits of detection in order to derive an upper bound for the radionuclide inventories in the Great Lakes, or in any environmental compartment within the lakes.

4.2 Need for a Reassessment of Environmental Monitoring of Nuclear Facilities to Support the Great Lakes Water Quality Agreement

The comments on monitoring in the previous four items are generic and do address specific data problems associated with individual facilities in each country. These comments led the Task Force to conclude that:

(3) There is a strong need for a comprehensive review of all monitoring activities at nuclear facilities with a view toward making the monitoring more accommodating to the needs of the Great Lakes Water Quality Agreement.

(4) Since there are policy and fiscal implications to any likely expansion or adjustment of the monitoring efforts, the Task Force calls upon the relevant atomic energy and environmental agencies in each country to explore in great detail the kinds of monitoring needed and the changes to the current protocols.

4.3 Reporting

The Task Force concludes that:

(5) There are significant differences in the scope of data reporting and analysis of United States and Canadian nuclear power plant emissions.

(6) The monitoring for toxic chemicals used in large quantities at nuclear power plants needs to be included in analyses of their impact on the Great Lakes ecosystem

(7) The monitoring of radionuclides does not include the identification of radioactive forms of toxic chemicals.

(8) The details of United States data reporting are greater and more helpful for the purpose of ecosystem impact analysis than is Canadian reporting, but the United States data come in mixed formats, which make them difficult to organize.

(9) Facilities in the United States have historically aggregated their data on an annual basis, but that the contract to continue this aggregation task has apparently been discontinued as a cost savings measure.

(10) The current biological monitoring and reporting is neither consistent nor adequate for lakewide assessments.

(11) Developing inventories for specific isotopes in biological compartments was a difficult task because no common reporting format for production/presentation of biological data is used.

Discussions

Conclusions 5 through 11 describe problems associated with using the specific data from individual nuclear facilities and associated monitoring sites. The conclusions address the scope of data collections, the completeness of such data collections from specific sites and facilities, the methods of reporting and aggregation of the data, and the problems of handling data from variable formats. In addressing these specific data issues, the Task Force noted the following important considerations:

(A) Since all of the Canadian nuclear power plant facilities belong to one corporate entity, the Canadian data are quite uniform in their scope, reporting, and formats. The United States nuclear power plant facilities, however, belong to some 15 different corporate entities. Thus, while the United States facilities report data that meet the requirements set by the (US) Nuclear Regulatory Commission, these data often vary in scope, reporting, and formats.

(B) To bring some semblance of order to the data from United States nuclear power plants, the Nuclear Regulatory Commission had previously contracted with the Brookhaven National Laboratories to produce an annual document, which assembled in a standardized format the emissions data from United States nuclear power plants. These reports often appeared 3 years after the individual facilities reported their emissions for a given year and usually reflected the varying timetables and lag times in the submissions of data from the United States facilities. The termination of the Brookhaven contract in 1996 without a new contractual effort represents a serious reporting setback for those groups interested in the radionuclide emissions from United States nuclear power facilities.

(C) The Task Force noted that the United States reporting tends to include a far greater number of radionuclides than the Canadian reporting, although the Task Force could not always judge whether the more extensive reporting by United States sources is more comprehensive and useful than the Canadian reporting. The United States data often report radionuclides at extremely low levels, basically limits of detection. The uncertainties in the reported data may call into question the information value of reporting selected radionuclides in certain emissions at levels of detection. On the other hand, the aggregated reporting of these radionuclides at trace levels does reveal much about the performance of the nuclear reactors and allows for a better understanding of the relationship between a particular reactor technology and the generation of its nuclear waste products.

(D) Biological data have multiple problems, ranging from sample descriptions to variable lower limits of the levels of detection of radionuclides. The latter problem has particularly troubled the Task Force, because for many radionuclides the lower limit changes with every sample even when the methodology and instrumentation do not change. This rather curious situation arises because of the need to back calculate and correct radionuclide data to the original time of sampling. Radioactivity continues to decay in a sample after collection and through the period of storage, analysis, and reporting. To place all measurements on a common basis, the radionuclide levels must be corrected to those at the time of sample collection.

(E) The large-scale use of nonradioactive toxic chemicals at nuclear power plants is often overlooked in establishing toxic substances inventories and monitoring activities. Among the chemical problems are those related to weed control on roadways and fence areas in a facility and at its perimeter, calling for considerable use of herbicides and pesticides. Facilities with cooling towers require the use of antifouling agents, water softening agents, and a variety chemicals to maintain heat transfer surfaces at their highest heat exchange capacities. The corrosion and fouling of piping and cooling system components, including water intakes, has led to widespread use of anti-corrosion and fouling-control agents. The problems of the zebra mussel has led to increased use of chlorine as a decontaminating agent. How these chemicals behave in contact with radioactivity is not assessed in any of the monitoring work.

4.4 Harmonization of Monitoring and Data Reporting

The Task Force concludes that:

(12) There is a need to harmonize the approaches used in the United States and Canada with respect to the scope of monitoring, the radionuclides reported, and the reporting of biological data. International cooperation among the nuclear agencies of both countries would accomplish much of this harmonization.

4.5 Biological Transfer Factors for Lake Systems

The Task Force concludes that:

(13) There is a special issue of reporting nuclear data, which applies specifically to the Great Lakes and has the implication of rendering incorrect some dose-assessment factors used in establishing the transfer of radionuclides from biota to humans in the region of interest. The issue relates to the transfer factors which estimate uptake of radionuclides in biota. These transfer factors traditionally have been derived from work done in rivers and oceans, rather than in freshwater lakes. The Task Force is concerned that the factors derived from riverine and oceanic systems are inappropriate for use in the Great Lakes.

Discussion

In developing the inventory for radionuclides, the Task Force noted that the bioaccumulation, biomagnification, and transfer factors used to describe the cycling of radionuclides and their transfer along exposure pathways to biota, including humans, came from the long history of work done in the marine, estuarine, and river environments. This work stemmed from interests in the deposition of radionuclides in the oceans and the transport of radionuclides down rivers and estuaries from discharges to the oceans. The comparable studies for lakes were virtually non-existent. Yet for the Great Lakes, the need for transfer factors that describe lake environments is critical.

To what extent can one use riverine, estuarine, or oceanic data to infer lake situations for the cycling and transfer of radionuclides in environmental compartments? Where no data exist, it is the obvious approach.

But why bother to use marine data when lake data exist that can be used to develop the appropriate factors? The Task Force undertook such analysis after discovering the nuclear sciences literature was not extensive in its coverage of lake situations.

To those persons who believe that the oceanic work, excellent as it was, should be used for the Great Lakes without confirmation, the Task Force cites two examples: radionuclides of silver, specifically ^{110,110m}Ag, and radionuclides of lanthanide elements (rare earths). These radionuclides appear in the effluents of nuclear power plants from the Great Lakes.

Silver, in the presence of chloride (the main anionic constituent of estuaries and oceans) forms silver chloride (AgCl), a compound with such a low water solubility in water that it is a basis for the quantitative analysis of silver. To reverse the solubility requires a large quantity of either ammonia or cyanide ion, such levels in environment being toxic in their own right. Because of nitrogen limitations of marine and estuarine environments, ammonia would not be present in these environments unless a specific pollutant source were present or an unusual algal species dominated plankton production. In lakes and rivers, however, where chloride is low and nitrogen is rarely limited, the presence of the silver radionuclides in soluble ionic form is almost a given. Only soluble silver is subject to biouptake, and biouptake factors for silver in freshwater systems are as high as 100,000. However, factors for silver do not exist for freshwater biota, and thus the marine factors are the ones in use.

Rare earth elements (the lanthanides) have unusual biological uptake. Freshwater organisms can often selectively accumulate these elements, and except for yttrium, cerium, and lanthanum, and in a few instances europium, usually only the even atomic numbered elements accumulate in freshwater biota. Thus, it is not correct to assume that all lanthanides accumulate and to use the marine factors, which rarely discriminate among lanthanides, but rather use cerium and lanthanum as surrogates for all of the elements in this group.

4.6 Radionuclides of Concern

Based on the Task Force's studies, it concludes that:

(14) There are radionuclides that merit separate studies and further reporting because of the patterns of use and discharge; physical, chemical, and biological properties; and the special monitoring needs of lakes as opposed to estuaries, oceans, and rivers (these include tritium, carbon-14, iodine-129, isotopes of plutonium, and radium-226).

(15) There are other radionuclides that could be a potential concern in special situations: technetium 99, -99m; phosphorus-32; chromium-51; cesium-134, -137; cerium-141, -144; strontium-89, -90; iodine-125, -131; and cobalt-60.

Discussions

The radionuclides listed in conclusion 14 are those which have exceptionally long half-lives, arise from both natural (cosmogenic and primordial) sources and some aspect of the nuclear fuel cycle, and present long-term toxicological and ecological problems. Except for ^{14}C and ^{129}I, the isotopes are routinely monitored in the Great Lakes.

The isotopes listed in conclusion 15 occur often in the discharges of sources other than nuclear power plants as well as in some cases in various components of the nuclear fuel cycle. Under conditions of large-scale emission or abundance, they merit special monitoring studies.

Murray Clamen

Murray Clamen,
Canadian Co-Chair

Joel L Fisher

Joel Fisher,
U.S. Co-Chair

Rosalie Bertell

Rosalie Bertell

Walter E. Carey

Walter Carey

Robert Krauel

John L Clark

John Clark

Bliss L. Tracy

Bliss Tracy

APPENDIX I

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APPENDIX II

Glossary

Absorbed dose: Energy deposited per unit mass when ionizing radiation passes through matter.

Accelerator: A device for increasing the velocity and energy of charged elementary particles, for example, electrons or protons, through application of electrical and/or magnetic forces. Accelerators have made particles move at velocities approaching the speed of light.

Actinides: A group of 15 elements with atomic number from 89 to 103 inclusive. All are radioactive and include thorium, uranium, plutonium, and amercium.

Activation: The process of making a material radioactive by bombardment with neutrons, protons, or other nuclear particles. Also called radioactivation.

Activity: The rate at which spontaneous transformations occur in the nuclei of a collection of atoms. The fundamental unit is the becquerel (Bq). One Bq is equal to one transformation per second. A traditional unit, still in use, is the curie (Ci). One Ci is equal to 3.7×10^{10} or 37,000,000,000 transformations per second.

Alpha particle (symbol α): A collection of four, primary nuclear particles, two protons and two neutrons. Its composition is identical to the nucleus of a helium atom. Its expulsion from a nucleus is one of several methods for accomplishing spontaneous nuclear transformation (also called "decay" or "disintegration"). It is the least penetrating of the three types of radiation commonly emitted from radioactive material has entered a living organism.

Anthropogenic: Made by humans.

Artificial radioactivity: Man-made radioactivity produced by fission, fusion, particle bombardment, or electromagnetic irradiation.

Atom: A particle of matter indivisible by chemical means. It is the fundamental building block of the chemical elements. Atoms are electrically neutral.

Background radiation: The radiation naturally present in the environment. It includes both cosmic rays and radiation from naturally occurring radioactive materials contained in the earth and in living organisms.

Becquerel (symbol Bq): See "Activity"

Beta particle (symbol β): A particle emitted from a nucleus during a certain type of spontaneous transformation. It is physically identical to an electron but may carry a positive or negative charge. It is somewhat more penetrating than an alpha particle but still represents a relatively minor risk when emitted from a source external to a living organism.

Bioaccumulation: The ability of an organism to build up concentrations of a chemical substance from its environment.

Biological half-life: The time required for a biological system, such as a human or an animal, to eliminate, by natural processes, half the amount of a substance that has entered it.

Biomagnification: The tendency for the concentrations of a chemical substance in living organisms to increase as one moves further up the food chain.

Biosphere: The biological components of the global ecosystems.

Biota: Living organisms.

Biouptake: The uptake of substances by living organism from the environment.

Body burden: The amount of radioactive material present in the body of a man or an animal.

Boiling water reactor: A reactor in which water, used as both coolant and moderator, is allowed to boil in the core. The resulting steam can be used directly to drive a turbine.

Breeder reactor: A reactor that produces fissionable fuel as well as consuming it, especially one that creates more than it consumes.

Cladding: The outer jacket of nuclear fuel elements. It prevents corrosion of the fuel and the release of fission products into the coolant. Aluminium or its alloys, stainless steel, and zirconium alloys are common cladding materials.

Collective Effective Dose: The quantity obtained by multiplying the average effective dose or committed effective dose by the number of persons exposed to a given radiation source (unit, person-sievert; symbol, person Sv).

Committed Effective Dose: The effective dose that will be accumulated over a period of time following a single intake of radioactive material into the body. Standard periods of integration are 50 years for adults and 70 years for a lifetime exposure.

Conversion: A step in the nuclear fuel cycle where uranium oxide (UO$_3$) is converted to uranium hexafluoride (UF$_6$), to be used in subsequent enrichment of the 235 isotope of uranium.

Coolant: A substance circulated through a nuclear reactor to remove or transfer heat. Common coolants are water, air, carbon dioxide, liquid sodium, and sodium–potassium alloy.

Cosmic rays: Radiation of many sorts but mostly atomic nuclei with very high energies, originating outside the earth's atmosphere. Cosmic radiation is part of the natural background radiation. Some cosmic rays are more energetic than any anthropogenic forms of radiation.

Cosmogenic: Secondary radionuclides produced by bombardment with primary cosmic rays, particularly in the upper atmosphere.

Curie: see "Activity"

Decay: The process of spontaneous transformation of a radionuclide. The decrease in the activity of a radioactive substance.

Decay Product: A radionuclide or radionuclide produced by decay. It may be formed directly from a radionuclide or as a result of a series of successive decays through several radionuclides.

Decommissioning: The act of removing a regulated facility from operation and operational regulation. This usually entails a certain amount of cleanup (decontamination).

Depleted uranium: Uranium having a smaller percentage of uranium-235 that the 0.7% found in natural uranium. It is obtained from the residues of uranium isotope separation.

Deuterium (symbol ^2H or D): An isotope of hydrogen whose nucleus contains one neutron and one proton and is therefore about twice as heavy as the nucleus of normal hydrogen, which is only a single proton. Deuterium is often referred to as heavy hydrogen; it occurs in nature as 1 atom to 6500 atoms of normal hydrogen. It is nonradioactive.

Diatoms: One-celled plants (algae) of the phylum Bacillerophyta that have a silicon shell (frustule) that remains as a fossil after the cell has died.

Dose: see "Absorbed Dose"

Ecosystem: The combination biological and non-biological interacting components that describe some system of interest.

Ecosystem Approach: The philosophy in the Great Lakes Water Quality Agreement that guides the development of processes, regulations, criteria, and objectives for programs and activities to meet the goals of the Agreement. The approach emphasizes the consideration of the effects and impacts of various activities and substances on all of the media and components of ecosystems in the Great Lakes.

Effective half-life: The time required for a radionuclide contained in a biological system, such as a man or an animal, to reduce its activity by half as a combined result of radioactive decay and biological elimination.

Electromagnetic radiation: Radiation consisting of associated and interacting electric and magnetic waves that travel at the speed of light. Examples are light, radio waves, gamma rays, and X-rays. All can be transmitted through a vacuum.

Electron volt (symbol eV): A unit of energy equivalent to the energy gained by an electron passing through a potential difference of one volt. It is used to quantify the amount of energy carried by all forms of ionizing radiation. Radiation emitted from nuclei during the process of spontaneous transformation generally have energies in the range of keV (= 1,000 eV) or MeV (= 1,000,000 eV)

Enrichment Factor: A ratio of activities or concentrations of an isotope in two media relative to the ratio of a standard chemical in those media.

Equivalent Dose: The quantity obtained by multiplying the absorbed dose by the appropriate radiation weighting factor to allow for the different effectiveness of the various ionizing radiations causing fatal cancers (unit, sievert; symbol, Sv).

Fallout: Air-borne particles containing radioactive material that fall to the ground following a nuclear explosion. "Local fallout" from nuclear detonations falls to the earth's surface within 24 hours after the detonation. "Tropospheric fallout" consists of material injected into the troposphere but not into the higher altitudes of the stratosphere. It does not fall out locally, but usually is deposited in relatively narrow bands around the earth at about the latitude of injection. "Stratospheric fallout" or "worldwide fallout" is that which is injected into the stratosphere and which then falls out relatively slowly over much of the earth's surface.

Fission: The splitting of a heavy nucleus into two approximately equal parts, accompanied by the release of a relatively large amount of energy and generally one or more neutrons. Fission can occur spontaneously, but usually is caused by nuclear absorption of gamma rays, neutrons, or other particles.

Fission products: The nuclei formed by the fission of heavy elements plus the radionuclides formed by the fission fragments' radioactive decay.

Foraminifera: Marine microinvertebrates that have a silicon shell, which remains as a fossil after the animal has died.

Fuel cycle: The series of steps involved in supplying fuel for nuclear power reactors. It includes mining, refining, the original fabrication of fuel elements, their use in a reactor, and radioactive waste disposal.

Gamma Rays (symbol γ): Photons of electromagnetic radiation, similar to X-rays. They are usually more energetic than X-rays and are emitted from nuclei during spontaneous transformation. Gamma rays are very penetrating and are best shielded against by dense material such as lead.

Genetic effects of radiation: Radiation effects that be transferred from parent to offspring. Any radiation-caused changes in the genetic material of sex cells.

Gray (symbol Gy): See "Absorbed dose"

Half-life (physical): The time in which half the atoms of a particular radioactive substance disintegrate. Measured half-lives vary from millionths of a second to billions of years.

Heavy water (symbol D_2O): Water containing significantly more than the natural proportion of heavy hydrogen atoms to ordinary hydrogen atoms. Heavy water is used as a moderator in some reactors, because it slows down neutrons effectively and also has a low cross section for absorption of neutrons.

Heavy-water-moderated reactor: A reactor that uses heavy water as its moderator. Heavy water is an excellent moderator and thus permits the use of inexpensive natural uranium as a fuel.

Homeostasis (homeostatic): A group of closely spaced physiological steady states for metabolic processes within which an organism, cell, tissue, or other biological components attempts to maintain itself for optimum performance.

Ion: An atom or molecule that has lost or gained one or more electrons. By this ionization it becomes electrically charged. Examples are an alpha particle, which is a helium atom minus two electrons, and a proton, which is a hydrogen atom minus its electron.

Ion exchange: A chemical process involving the reversible interchange of various ions between a solution and a solid material, usually a plastic or a resin. It is used to separate and purify chemicals, such as fission products or rare earths, in solutions.

Ionization: The process of adding one or more electrons to, or removing one or more electrons from, atoms or molecules, thereby creating ions. High temperatures, electrical discharges, or nuclear radiations can cause ionization.

Ionizing radiation: Radiation capable of dislodging one or more electrons from atoms or molecules, thereby producing ions. Examples are alpha particles, beta particles, X-rays, and gamma rays.

Isotope: Differing forms of a particular chemical element. The atoms of all forms will have the same number of protons in each nucleus and the same number of electrons surrounding the nucleus. Hence, the chemical behaviour of all forms will be essentially identical. However, each version's nuclei will have a number of neutrons that is different from any other version. Thus, the isotopes (forms) of a particular element will have different physical properties, including the mass of its atoms and whether the nuclear structure of its atoms will retain its identity indefinitely (be "stable") or undergo spontaneous transformation at some future time (be "radioactive").

Limnology: The study of inland waters, lakes, and rivers.

Lower Limit of Detection (symbol LLD): This is the lowest concentration of radioactive material in a sample that can be detected at the 95% confidence level with a given analytical system.

Macrophyte: Rooted aquatic plants.

Moderator: A material, such as ordinary water, heavy water, or graphite, used in a reactor to slow down high-velocity neutrons, thus increasing the likelihood of further fission.

Natural radioactivity: The property of radioactivity exhibited by more than 50 naturally occurring radionuclides.

Neutron: An uncharged primary nuclear particle with a mass slightly greater than the other primary nuclear particle, the proton. When emitted from a nucleus following a fission or other nuclear event, a neutron can cause ionization indirectly. A "free" neutron, normally present in our environment, is unstable and exhibits a half life of about 13 minutes.

Non-ionizing radiation: Radiation that is not capable of dislodging electrons from atoms or molecules (see ionizing radiation). Examples of non-ionizing radiation are radio waves, microwaves, and light.

Nuclear Reactor: A device in which a fission chain reaction can be initiated, maintained, and controlled. Its essential component is a core with fissionable fuel. It usually has a moderator, a reflector, shielding, coolant, and control mechanisms.

Nuclear Fission: The process in which a nucleus splits into two or more nuclei and energy is released.

Nucleus: The small, positively charged core of an atom. It is only about $^1/_{10,000}$ the diameter of the atom but contains nearly all the atom's mass. All nuclei contain both protons and neutrons, except the nucleus of ordinary hydrogen, which consists of a single proton.

Radionuclide: A species of atom characterized by the constitution of its nucleus, which is specified by its atomic mass and atomic number (Z), or by its number of protons (Z), number of neutrons (N), and energy content.

Plutonium (symbol Pu): A heavy, metallic element with atomic number 94. An important isotope is Pu-239 produced by neutron irradiation of uranium-238. Plutonium is used in reactor fuel, weapons, and specialized mobile power sources such as space probes and heart pacemakers.

Primordial: Present since the beginning of the earth.

Proton: An elementary particle with a single positive electrical charge and a mass approximately 1837 times that of the electron. The nucleus of an ordinary or light hydrogen atom. Protons are constituents of all nuclei. The atomic number (Z) of an atom is equal to the number of protons in its nucleus.

Quality factor (QF): A factor by which an absorbed dose is multiplied to more closely correspond to a biological effect produced. Dose (Gy) × QF = Dose Equivalent (Sv).

Rad: The traditional unit of absorbed dose of ionizing radiation. A dose of one rad results from the absorption of 100 ergs of energy per gram of material. It has been supplanted by the gray (Gy) which is equal to one joule/kilogram.

Radiation: The emission and propagation of energy through matter or space by means of electromagnetic disturbances, which display both wave-like and particle-like behaviour; in this context the "particles" are known as photons. Also, the energy so propagated. The term has been extended to include streams of fast-moving particles. Nuclear radiation is that emitted from atomic nuclei in various nuclear reactions, including alpha, beta, and gamma radiation and neutrons.

Radioactive Fallout: Radioactive materials deposited from the atmosphere.

Radioactive series: A succession of radionuclides, each of which transforms by radioactive disintegration into the next until a stable radionuclide results. The first member is called the parent, the intermediate members are called progeny, and the final stable member is called the end product.

Radioactive decay: The spontaneous transformation of one radionuclide into a different radionuclide or into a different energy state of the same radionuclide. The process results in a decrease, with time, of the number of the original radioactive atoms in a sample. It involves the emission from the nucleus of alpha particles, beta particles, or gamma rays; as the nuclear capture or ejection of orbital electrons; or fission.

Radioactivity: See "Activity."

Radioisotope: An unstable isotope (version) of a chemical element. Nuclei of atoms of such an isotope undergo spontaneous transformation and emit radiation.

Radium (symbol Ra): A radioactive metallic element with atomic number 88. As found in nature, the most common isotope has an atomic weight of 226. It occurs in minute quantities associated with uranium in pitchblende, carnotite, and other minerals; the uranium decays to radium in a series of alpha and beta emissions. By virtue of being an alpha- and gamma-emitter, radium is used as a source of luminescence and as a radiation source in medicine and radiography.

Radon (symbol Rn): A radioactive element, the heaviest of the noble or inert gases known. Its atomic number is 86, and it atomic weight is 222. It is a progeny of radium in the uranium radioactive series.

Rem: A traditional unit of dose equivalent obtained by multiplying the absorbed dose in rad by the appropriate quality factor and any other necessary modifying factors. The rem has been superseded by the seivert (Sv).

Reprocessing: The extraction of useful fissionable materials from spent reactive fuel rods.

SI units: The International System of Units as defined by the General Conference of Weights and Measures in 1960. These units are generally based on the metre/kilogram/second units, with special quantities for radiation including the becquerel, gray, and sievert.

Sievert: The SI unit of dose equivalent. It is equal to the absorbed dose in gray (Gy) times the appropriate quality factor (QF) times any other necessary modifying factors. One sievert (Sv) is equal to 100 rem.

Spallation: The splitting off of small fragments from a nucleus under bombardment by high-energy particles.

Spontaneous Fission: The spontaneous splitting of a nucleus.

Stable Nuclei: The nuclei of a non-radioactive substance.

Tailings: Waste material remaining after a useful substance has been extracted from a mineral ore.

Technologically enhanced: Refers to a situation where the concentrations of naturally-occurring radionuclides, in environmental media are increased as a result of human activities, e.g., the mining and milling of mineral ores containing radioactive substances.

Terrestrial Sources: Natural radiation sources in the earth's crust or biosphere, as opposed to sources from space.

Thorium series (sequence): The series of radionuclides resulting from the radioactive decay of thorium-232. Many anthropogenic radionuclides decay into this sequence. The end product of this sequence in nature is lead-208.

Thorium (symbol Th): A naturally radioactive element with atomic number 90 and, as found in nature, an atomic weights of approximately 232. The thorium-232 isotope is abundant and can be transmitted to fissionable uranium-233 by neutron irradiation.

Transfer Factor: The ratio at equilibrium of the concentration of a chemical substance in one biological compartment to that in another biological compartment.

Transuranic element: An element beyond uranium in the Periodic Table, that is, with an atomic number greater than 92. All 11 transuranic elements are produced artificially and are radioactive. They are neptunium, plutonium, americium, curium, berkelium, californium, einsteinium, fermium, mendelevium, nobelium, and lawrencium.

Tritium: A radioactive isotope of hydrogen with two neutrons and one proton in the nucleus. It is both naturally occurring and produced in nuclear reactors. It is used in weapons, biomedical research, and in self-illuminating devices. It has a physical half-life of 12.33 years. See tritium terminology Text Box.

Uranium Series: The series of radionuclides resulting from the radioactive decay of uranium-238, also known as the uranium–radium series. The end product of the series is lead-206. Many anthropogenic radionuclides decay into this sequence.

Uranium (symbol U): A radioactive element with the atomic number 92 and, as found in natural ores, an average atomic weight of approximately 238. The two principal natural isotopes are uranium-235 (0.7% of natural uranium), which is fissionable, and uranium-238 (99.3% of natural uranium). Natural uranium also includes a minute amount or uranium-234. Uranium is the basic raw material of nuclear energy.

X-rays: A penetrating form of electromagnetic radiation emitted either when the orbital electrons of an excited atom return to their normal state or when a target is bombarded with high-speed electrons. X-rays are always non-nuclear in origin.

APPENDIX III

Acronyms and Abbreviations

AECB: Atomic Energy Control Board (Canada)

BEIR: National Research Council's Committees on Biological Effects of Ionizing Radiation (United States)

BWR: Boiling Water Reactor

CAMECO: CAnadian Mining and Energy Company

CANDU: CANadian Deuterium Uranium refers to the Canadian design of a nuclear power reactor, which utilizes natural uranium fuel and heavy water moderator.

DOE: Department of Energy (United States)

EPA: Environmental Protection Agency (United States)

FBR: Fast-Breeder Reactor

GCR: Gas-Cooled Reactor

GLWQB: Great Lakes Water Quality Board

HTO: tritiated water (Hydrogen – Tritium – Oxygen)

HWR: Heavy Water Reactor

IAEA: International Atomic Energy Agency

ICRP: International Commission for Radiation Protection

IJC: International Joint Commission

LLD: Lower Limit of Detection

LWR: Light Water Reactor

NCRP: National Council On Radiation Protection (United States)

NRC: Nuclear Regulatory Commission (United States)

NYSERDA: New York State Energy Research and Development Agency

PWR: Pressurized Water Reactor

QF: Quality Factor

TBT: Tissue Bound Tritium

TENR: Technologically Enhanced Natural Radioactivity

UNSCEAR: United National Scientific Committee on the Effects of Atomic Radiation publishes periodic reports on sources and effects of ionizing radiation.

Elements

Ac, actinium; Ag, silver; Al, aluminum; Am, americium; Ar, argon;
B, boron; Ba, barium; Be, beryllium; Bi, bismuth; Br, bromine;
C, carbon; Ca, calcium; Cd, cadmium; Ce, cerium; Cl, chlorine; Co, cobalt; Cr, chromium; Cs, cesium; Cu, copper;
F, fluorine; Fe, iron; Fr, francium;
Gd, gadolinium;
H, hydrogen; He, helium; Hf, hafnium;
I, iodine; In, indium;
K, potassium; Kr, krypton;
La, lanthanum; Lu, lutetium;
Mg, magnesium; Mn, manganese; Mo, molybdenum;
N, nitrogen; Na, sodium; Nb, niobium; Nd, neodymium; Np, neptunium;
O, oxygen;
P, phosphorus; Pa, protractinium; Pb, lead; Pd, palladium; Po, polonium; Pt, platinum; Pu, plutonium;
Ra, radium; Rb, rubidium; Re, rhenium; Rn, radon; Ru, ruthenium;
S, sulfur; Sb, antimony; Sc, scandium; Se, selenium; Si, silicon; Sm, samarium; Sn, tin; Sr, strontium;
Ta, tantalum; Tc, technetium; Te, tellurium; Th, thorium; Tl, thalium;
U, uranium;
V, vanadium;
Xe, xenon;
Y, yttrium; Yb, ytterbium;
Zn, zinc; Zr, zirconium.

Units of Measurement

Bq, becquerel; mBq, millibecquerel (10^{-3} Bq); kBq, kilobecquerel (10^{3} Bq); MBq, megabecquerel (10^{6} Bq); GBq, gigabecquerel (10^{9} Bq); TBq, terabecquerel (10^{12} Bq); PBq, petabecquerel (10^{15} Bq);

Ci, curie; nCi, nanocurie (10^{-9} Ci); µCi, microcurie (10^{-6} Ci); MCi, megacuries (10^{6} Ci)

dpm, disintegrations per minute

eV, electron volts; keV; kiloelectron volt (10^{3} eV) ; MeV, megaelectron volt (10^{6} eV)

g, gram; µg, microgram (10^{-6} g); mg, milligram (10^{-3} g); kg, kilogram (10^{3} g)

Gy, gray

ha, hectare

hr, hour

L, litre; mL, millilitre (10^{-3} L)

m, metre; cm, centimetre (10^{-2} m); km, kilometre (10^{3} m)

s, second

Sv, sievert; mSv, millisievert (10^{-3} Sv)

V, volts

W, watt; MW, megawatt (10^{6} W); GW, gigawatt (10^{9} W)

APPENDIX IV

Terms of Reference: Nuclear Task Force

The International Joint Commission (Commission) has authorized a "Nuclear Task Force" to review, assess and report on the state of radioactivity in the Great Lakes and to carry out such other activities as the Commission may, in future, so direct.

(1) The name of the Task Force shall be the Nuclear Task Force.

(2) The Task Force shall undertake, as its initial project, a review and assessment of the status of radioactivity in the Great Lakes. This project is to be completed in connection with the current biennial Great Lakes reporting cycle. A report of this work shall be available along with the other reports at the Great Lakes Biennial Meeting. The Commission may or may not require that similar reports be prepared for subsequent Great Lakes Biennial Meetings.

(3) The Task Force shall propose such additional projects as it deems important, based in part on the following criteria:

   (a) Work performed on the State of Radioactivity in the Great Lakes Report. This report shall be the principal vehicle upon which to base recommendations for projects since the report has as one of its production objectives the prioritization of nuclear problems in the Great Lakes requiring analysis and remediation.

   (b) Concerns of the Commissioners.

   (c) Problems brought to the attention of the Task Force by its Members, Associates, and others in the course of its work.

(4) The Task Force shall make its first official recommendation of additional projects upon the completion of work for its first assessment of the state of radioactivity in the lakes.

(5) The Nuclear Task Force shall undertake such other projects as the Commission directs.

(6) The Task Force shall seek from the Commission directly whatever resources and funds are needed for specific projects and support.

(7) The Task Force shall consist of, at most, six 'Members' chosen from staff of the the United States and Canadian Sections and of the Regional Office and from the Water Quality Board, the Great Lakes Science Advisory Board, and the Council of Great Lakes Research Managers. In addition, the Task Force shall utilize the services of 'Associates', who are specialists in a variety of nuclear issues and who can provide assistance, support, analysis, information, data and appropriate liaison to groups outside of the Commission. There are no restrictions on the number of Associates. All members and Associates shall receive letters of appointment which specify their status and responsibilities.

(8) Members shall direct the work of the Task Force, be responsible for the production of its reports, and for the presentation of reports and views of the Task Force. Associates shall support the work of the Task Force, but will not ordinarily be called upon to present their views or the views of the Task Force to the Commission.

(9) The Commission shall appoint two co-chairs of the Nuclear Task Force and will approve associate members.

(10) The Task Force may establish whatever subgroups it deems necessary for its work. A subgroup shall be chaired by a Member.

(11) The Task Force shall report directly to the Commission.

(12) The Task Force will be sunsetted in five years.

(13) The start date of the Task Force was December 1994.

APPENDIX V

Membership of the Nuclear Task Force

DR. MURRAY CLAMEN, CO-CHAIR, CANADIAN SECTION, INTERNATIONAL JOINT COMMISSION
Dr. Clamen is Secretary of the Canadian Section of the International Joint Commission (IJC) and is responsible for the day-to-day operations and management of the Section. A registered professional engineer, his expertise is in international water resource studies and environmental assessments. His career has included experience in the private sector with consulting engineering and research firms in Quebec and British Columbia and a total of 20 years in the Federal Public Service; 17 years with the IJC, and three with Environment Canada. At the IJC he has provided technical and policy advice to the Commissioners on a wide range of transboundary issues and participated in numerous Canada/U.S. studies and assessments.

DR. JOEL FISHER, CO-CHAIR, US SECTION, INTERNATIONAL JOINT COMMISSION
Dr. Fisher's environmental work began in the military service where he worked on programs to dissemble, disarm and dispose of nuclear and chemical munitions. Later at the United States Environmental Protection Agency he worked for several years on programs which addressed the environmental fate and behaviour of pollutants in the emissions from fossil fuel and nuclear power plants. At the International Joint Commission, he advises on the problems of environmental fate and behaviour of pollutants which have transboundary implications.

DR. ROSALIE BERTELL, GNSH, INTERNATIONAL INSTITUTE OF CONCERN FOR PUBLIC HEALTH
Dr. Bertell has worked professionally in Environmental Epidemiology since 1968, serves on the Advisory Boards for the Great Lakes Health Effects Program of Health Canada, and the Ontario Environmental Assessment Board and has been a member of the IJC Science Advisory Board. She has published a *Handbook for Estimating the Health Effects of Exposure to Ionizing Radiation* and the popular non-fiction book: *No Immediate Danger: Prognosis for a Radioactive Earth*, together with more than 100 other publications.

DR. BLISS TRACY, HEALTH CANADA
Dr. Tracy heads a group on radiological impact at the Radiation Protection Bureau. He has carried out a number of research projects on environmental radioactivity including the uptake of radiocesium in Arctic food chains, uranium uptake and metabolism in humans, and radioactivity in Great Lakes ecosystems. Dr. Tracy provides advice regularly toward environmental impact assessments of nuclear and radioactive waste disposal facilities. He is also involved in planning for the Federal Nuclear Emergency Plan and is contributing to the design of an international monitoring system for verification of the Comprehensive Test Ban Treaty.

MR. ROBERT KRAUEL, ENVIRONMENT CANADA
Mr. Krauel is manager of the Environmental Contaminants and Nuclear Programs Division in Environment Canada's Ontario Regional Office. Mr. Krauel has been Coordinator of the Federal - Provincial Remedial Action Plan Team for Port Hope Harbour. He has participated in several environmental assessment reviews related to the nuclear industry, including uranium mine decommissioning, low level radioactive waste management, and uranium refining and conversion.

DR. WALTER CAREY, OHIO STATE UNIVERSITY
Dr. Carey is a private Consultant and a Certified Health Physicist. During 35 years at the Ohio State University, he taught courses on Zoology and Nuclear Engineering. He also served as the original Director of the Nuclear Reactor Laboratory and later as the University's Radiation Council, and is the Chair of the Council's Radiation Protection Standards Committee. He is a Diplomat of the American Academy of Health Physics, a member of Sigma Xi and an Emeritus Member of the American Nuclear Society.

DR. JOHN CLARK, REGIONAL OFFICE, INTERNATIONAL JOINT COMMISSION
Dr. Clark received a doctorate in Environmental Health from the University of Cincinnati in 1970. Prior to joining the Commission's staff in 1974 he served as a Public Health Analyst and as a Statistician with the United States Public Health Service.