Radionuclides in the Great Lakes Basin

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The Great Lakes basin is of radiologic interest due to the large population within its boundaries that may be exposed to various sources of ionizing radiation. Specific radionuclides of interest in the basin arising from natural and artificial sources include ^{3}H, ^{14}C, ^{90}Sr, ^{129}I, ^{131}I, ^{137}Cs, ^{222}Rn, ^{226}Ra, ^{235}U, ^{238}U, ^{239}Pu, and ^{241}Am. The greatest contribution to total radiation exposure is the natural background radiation that provides an average dose of about 2.6 mSv/year to all basin residents. Global fallout from atmospheric nuclear weapons tests conducted before 1963 has resulted in the largest input of anthropogenic radioactivity into the lakes. Of increasing importance is the radionuclide input from the various components of the nuclear fuel cycle. Although the dose from these activities is currently very low, it is expected to increase if there is continued growth of the nuclear industry. In spite of strict regulations on design and operation of nuclear power facilities, the potential exists for a serious accident as a result of the large inventories of radionuclides contained in the reactor cores; however, these risks are several orders of magnitude less than the risks from other natural and man-made hazards. An area of major priority over the next few decades will be the management of the substantial amounts of radioactive waste generated by nuclear fuel cycle activities. Based on derived risk coefficients, the theoretical incidence of fatal and weighted nonfatal cancers and hereditary defects in the basin's population, attributable to 50 years of exposure to natural background radiation, is conservatively estimated to be of the order of 3.4×10^{5} cases. The total number of attributable health effects to the year 2050 from fallout radionuclides in the Great Lakes basin is of the order of 5.0×10^{3}. In contrast, estimates of attributable health effects from 50 years of exposure to current nuclear fuel cycle effluent in the basin are of the order of 2×10^{2}. Although these are hypothetical risks, they show that the radiologic impact of man-made sources is very small compared to the effects of normal background radiation. — Environ Health Perspect 103(Suppl 9):89–101 (1995)

Key words: dose, fallout, Great Lakes, nuclear power, radioactivity, risk

Introduction

Before 1942, human exposure to ionizing radiation was limited to natural radioactivity and medical diagnosis. In December 1942, the first controlled, self-sustaining nuclear chain reaction was achieved, followed in July 1945 by the first successful test of an atomic bomb. Since then, the uses of nuclear energy have become more diverse and widespread, encompassing medical diagnosis and treatment, nuclear power, and consumer and industrial applications. These applications, however, release radioactivity into the global ecosystem and have added to the levels of existing natural radiation, provoking concern over the possible health effects associated with increased radiation exposure.

Radionuclides present in the biosphere, whether natural or artificial in origin, ultimately result in irradiation of human populations. The biologic consequences of ionizing radiation exposure involve tissue damage and can cause immediate physiologic harm within a few days or weeks following a large, acute individual dose or delayed effects, the most important of which is the development of various cancers after an extended latent period following low, chronic exposures. Doses received from natural radioactivity and routine exposures from regulated practices are well below levels that would result in immediate harm.

The Great Lakes basin is an area of radiologic interest due to both actual and potential exposures that may be received by its large population. Comprising one of the world's largest sources of freshwater and supporting a population of over 36 million residents, the basin is unique in that it contains nearly all components of the nuclear fuel cycle, from uranium mining to radioactive waste management, as shown in Figure 1 (*1,2*). There are presently 16 operational nuclear generating stations located on the shores of Lakes Huron, Michigan, Erie, and Ontario, with a total installed electrical generating capacity of 27,000 megawatts (MW). As a result of the large inventories of radioactive material at these facilities, there is a potential for a significant accidental release of radionuclides into the environment. Although the probability of such an occurrence is extremely small, the health, social, and economic consequences could be significant; this requires consideration in radiologic assessments of the basin's environment. In addition, accidents at nuclear installations situated beyond the Great Lakes have the potential for contamination of the basin as a result of long-range atmospheric transport.

Sources

Natural Radioactivity

By far, the greatest contribution to the average public radiation exposure comes from radioactive elements in the earth's crust and from cosmic radiation originating in deep space. Natural sources contribute on average more than 98% of the human radiation dose, excluding medical exposures (*3*). The global average dose from natural sources as estimated by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) (*4*) is about 2.4 mSv/year, which can be compared with the National Council on Radiation Protection and Measurements estimate of 2.6 mSv/year for Canada (*5*). Exposure is both external, from direct cosmic and terrestrial radiation, and internal, from inhalation and ingestion of terrestrial and cosmogenic radionuclides found in air, water, food, and soil.

Terrestrial radiation exposure originates from the primordial radionuclides, whose half-lives are comparable to the age of the

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Abbreviations used: AECB, Atomic Energy Control Board, Canada; CANDU, Canada Deuterium Uranium; ICRP, International Commission on Radiological Protection; NCRP, National Council on Radiation Protection and Measurements; UNSCEAR, United Nations Scientific Committee on the Effects of Atomic Radiation; WHO, World Health Organization; Bq, becquerel; GW, gigawatt; MW, megawatt [MW(e) denotes electrical power output]; Sv, Sievert; $t_{1/2}$, radioactive half-life.

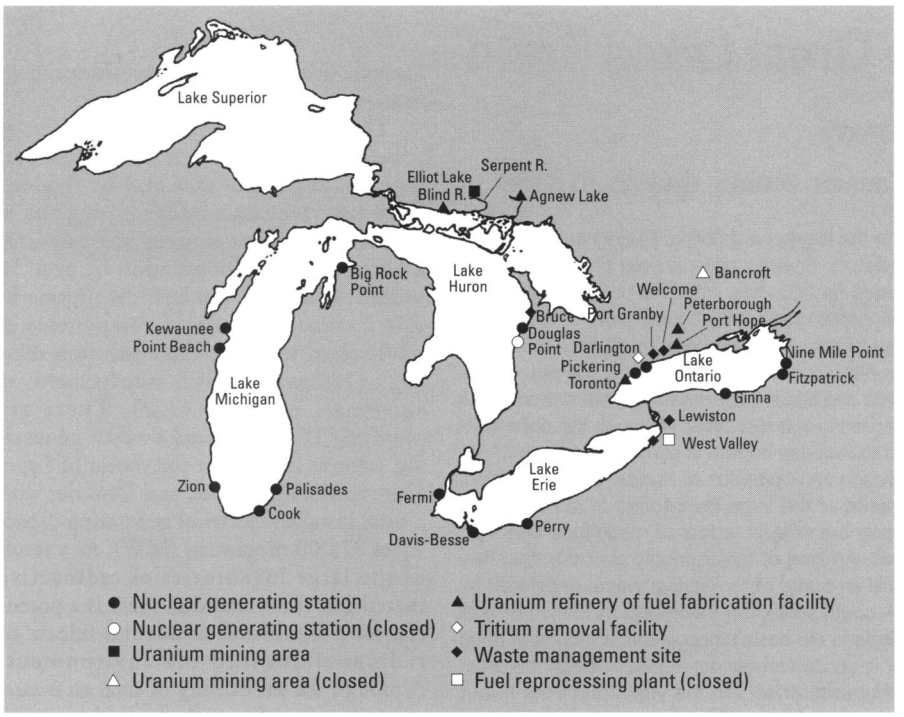

Figure 1. Nuclear fuel cycle facilities in the Great Lakes basin. Data from the International Joint Commission (*1*) and Joshi (*2*).

earth, and the secondary radionuclides produced by their radioactive decay. The naturally occurring radionuclides include mainly ^{40}K, and the three radioactive decay chains originating with ^{238}U, ^{232}Th, and ^{235}U. These radionuclides are ubiquitously present in low concentrations in soil and water as a result of weathering and erosion of rock. The isotopic abundance of ^{40}K in natural potassium is only about 0.012%, but because potassium is widespread and is taken into the body as an essential element, it contributes on average about one-tenth of the internal dose from natural radiation (*4*). Another major exposure pathway to natural radiation results from the decay of ^{226}Ra in the ^{238}U series. This decay results in the formation of gaseous ^{222}Rn, which can enter the atmosphere through emanation from soil and building materials. The principal sources of internal exposure, and a major component of total background radiation exposure, are the rapidly decaying radionuclides formed as a result of successive decays of ^{222}Rn. Exposure occurs when these radionuclides, namely ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po, are inhaled and retained in the lungs.

Additional but minor contributions to exposure come from the remaining nonseries primordial radionuclides, primarily ^{87}Rb (*5*), and cosmogenic radionuclides produced in the atmosphere by the interaction of cosmic rays with atmospheric argon, oxygen, and nitrogen. Cosmogenic radionuclides reach the earth through atmospheric mixing, precipitation scavenging, and gravitational settling; exposures result primarily from ingestion and are relatively constant throughout the world. The four radionuclides that contribute a measurable dose to humans are ^{14}C, ^{3}H, ^{22}Na, and ^{7}Be, on the order of 12 μSv annually, but the greatest contribution to this dose is from ^{14}C since it is a relatively long-lived radionuclide and a major constituent in body tissue (*4*).

The annual average contribution due to all internally deposited radionuclides is approximately 1.6 mSv, of which about 1.1 mSv results from the inhaled radon decay products. Actual individual exposures to background radioactivity in air, food, and water are, however, highly variable and depend on numerous factors including the amount, type, and availability of the radionuclide in the environment and the amount inhaled or ingested by the individual. Average exposure of the population in Canada and the United States from various sources of natural radiation is provided in Table 1 (*5*).

Atmospheric Weapons Fallout

In addition to the natural background radiation, nuclear technologies over the past 50 years have introduced significant quantities of artificial radionuclides into the global environment. The majority of these radionuclides have come from atmospheric nuclear weapons tests conducted in the 10-year period immediately before the 1963 limited test ban treaty, although additional tests have taken place since then by nonsignatory nations. Radionuclides produced by these tests have been distributed globally, with the maximum time-integrated weapons fallout per unit area occurring in the North Temperate Zone (40°–50°N), which encompasses the Great Lakes basin. Weapons fallout is the principal source of radionuclides in the basin. Although continued underground weapons testing has resulted in occasional venting of radioactive material to the atmosphere, the impact of these tests on environmental fallout levels has been insignificant (*4*).

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, having been measured in air, water, soil, and food products. Other radionuclides include ^{95}Zr, ^{95}Nb, ^{106}Ru, ^{131}I, ^{144}Ce, $^{239,240}Pu$, ^{241}Pu, and ^{241}Am. Estimates of

Table 1. Summary of total effective dose rates from various sources of natural background radiation in Canada and the United States.

	Total effective dose rate, mSv/year					
Source	Lung	Gonads	Bone surfaces	Bone marrow	Other tissue	Total
Cosmic radiation	0.03	0.07	0.008	0.03	0.13	0.27
Cosmogenic nuclides	0.001	0.002	—	0.004	0.003	0.01
External terrestrial	0.03	0.07	0.008	0.03	0.14	0.28
Inhaled nuclides	2.0	—	—	—	—	2.0
Nuclides in body	0.04	0.09	0.03	0.06	0.17	0.40
Totals (rounded)	2.1	0.23	0.05	0.12	0.44	3.0[a]

[a]The effective dose rates for Canada are about 20% lower for the terrestrial and inhaled components; the average effective dose rate in Canada from natural radiation is 2.6 mSv/year. Data from the National Council on Radiation Protection and Measurements (*5*).

the 1983 inventory of fallout ^{3}H, ^{90}Sr, and ^{137}Cs in the lakes are shown in Table 2 (*2*). Annual measurements of gross beta radioactivity from fallout in air and precipitation at Canadian monitoring sites around the lakes are shown in Figure 2 (*6,7*). Measurements of fallout ^{90}Sr and ^{137}Cs in milk samples from regions around these sites are shown in Figure 3. These decreasing values are similar to the national averages for Canada.

The total dose that will be received by individuals in the North Temperate Zone during the first 100 years following the initiation of nuclear weapons testing, for all atmospheric detonations conducted between 1945 to 1980, is estimated to be about 1.9 mSv (*4*). Although this dose represents only 5% of the committed dose from long-lived ^{14}C, all other fallout radionuclides will have delivered almost all of their dose during this period. In addition, the truncated dose provides a measure of the radiation hazard presented to those living during the period of intensive testing before 1963 and is equivalent to slightly less than 1 extra year of exposure to natural background radiation.

Nuclear Fuel Cycle

Increases in local exposures above natural background levels may result from radionuclides released during the various stages of the nuclear fuel cycle. Nearly all components of the nuclear fuel cycle are found within the basin, the main elements of which are uranium mining, fuel preparation, reactor operations, fuel reprocessing, and waste management. Currently, fuel reprocessing is not conducted in the basin, although a facility was in operation at West Valley, New York, from 1966 to 1972.

Three categories of radionuclides are associated with the fuel cycle phases: ^{238}U and ^{232}Th decay series radionuclides that are released in uranium mining and milling operations, which enhance the levels of natural terrestrial radionuclides; radioactive fission products and actinides produced in the nuclear fuel during normal reactor operation; and those produced by neutron absorption in structural and fuel-cladding materials during reactor operation. The impact of normal fuel cycle effluent on the basin ecosystem is small; however, the consequence of a large-scale accidental discharge of radioactivity from an operating nuclear reactor or storage facility, though extremely unlikely, must be considered in a radiologic assessment of the basin.

Uranium Mining Activities. All uranium mining and milling activities in the Great Lakes basin are situated in Canada. At present, there are four active uranium mine, mill, and tailings management areas, all located in the Serpent River basin on the North Channel of Lake Huron in the Elliot Lake area (Figure 1). Eleven other mine and mill operations in the Serpent River, Espanola, and Bancroft regions of

Table 2. Inventory of radionuclides in the Great Lakes from fallout to 1983, nuclear facility releases, and 1989 inventories stored at the facilities.

Radionuclide	Estimated radionuclide inputs and inventories by lake, TBq				
	Superior	Michigan	Huron	Erie	Ontario
Tritium					
Fallout[a]	7×10^{4}	6×10^{4}	7×10^{4}	4×10^{4}	3×10^{4}
Nuclear facilities	—	2×10^{3}	1.5×10^{4}	2×10^{2}	5×10^{2}
Strontium-90					
Fallout[a]	123	98	98	45	33
Nuclear facilities	—	0.015	0.11	1.5	0.15
Stored at facilities	—	5×10^{6}	3.5×10^{6}	6×10^{5}	4×10^{6}
Cesium-137					
Fallout[a]	200	159	159	74	54
Nuclear facilities	—	9	0.12	0.2	25
Stored at facilities	—	8×10^{6}	5×10^{6}	7×10^{5}	7×10^{6}

TBq, terabecquerels. [a]Input from fallout was calculated by Joshi (*2*) using deposition flux at mid-basin location for each lake using New York City data and adjusting for latitude.

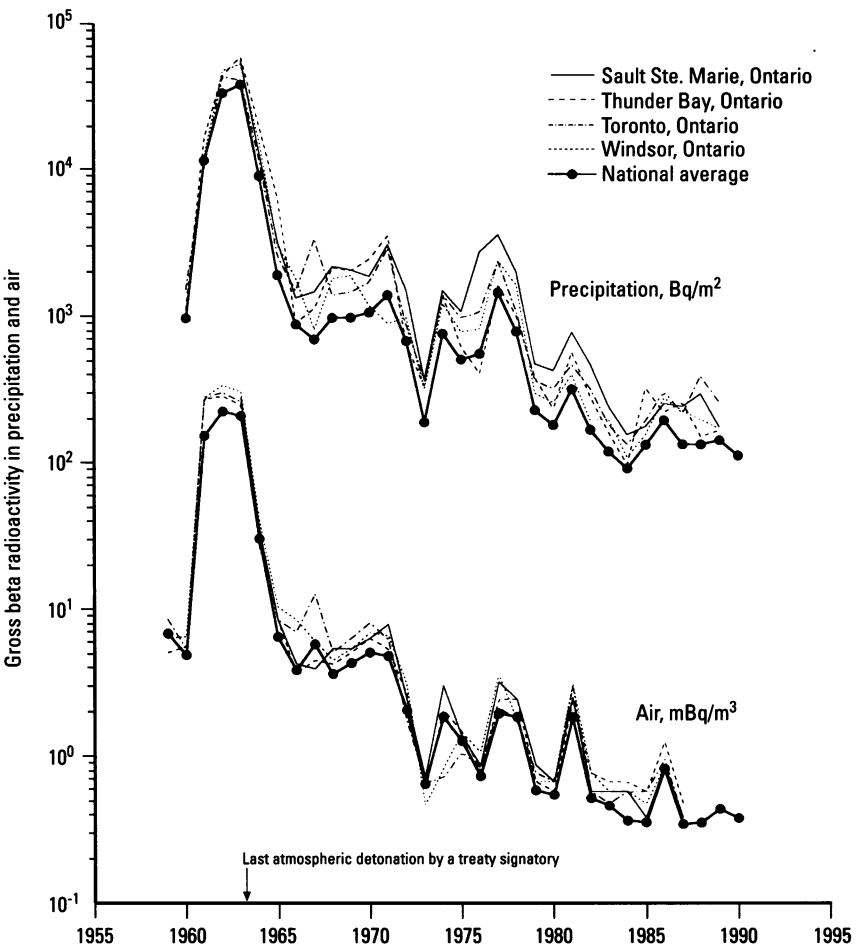

Figure 2. Gross beta radioactivity in precipitation and air. Annual averages at basin stations and national averages for Canada from 1959 to 1990. Data from Health Canada (*6*).

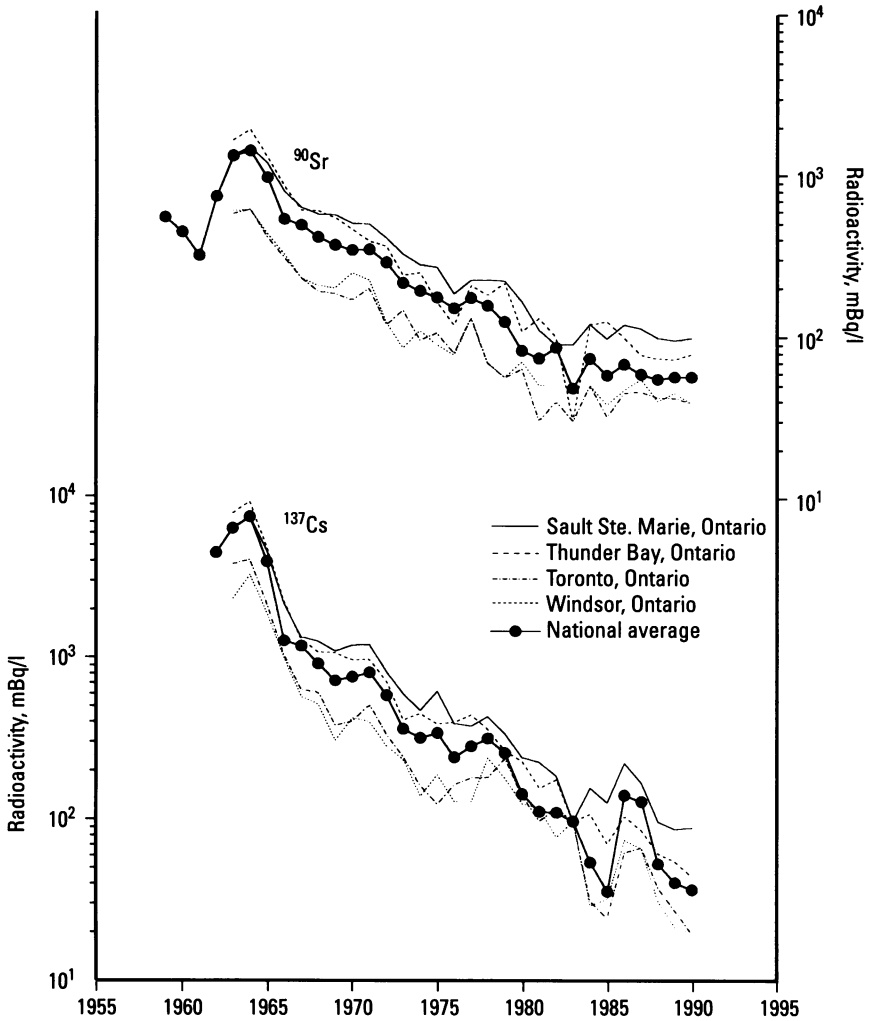

Figure 3. ^{90}Sr and ^{137}Cs radioactivity in milk samples. Annual averages from regions in the basin and national averages for Canada from 1959 to 1990. Data from Health Canada (*6*).

Ontario have been decommissioned, the majority of which have been revegetated or reclaimed (*8*).

Milling facilities receive mined uranium ores for conversion into U_3O_8 (yellowcake) concentrate for further processing. More than 95% of the uranium is removed in the milling process, along with approximately 10 to 15% of the radioactive material; the remaining 85 to 90% remains with the ore tailings as a long-term source of ^{226}Ra and ^{222}Rn gas (*9*). The long half-life of ^{226}Ra (1600 years) maintains the production of ^{222}Rn for thousands of years. Tailings piles are a potential source of radioactive contamination due to emanation of ^{222}Rn and to dispersion of the tailings by wind and water, which is determined by the degree of stabilization of the pile and the waste management procedures implemented.

Radon is the most important radionuclide released from mining activities. In terms of volume, mine and mill wastes have had the greatest impact on local ecosystems within the basin, despite their low radioactivity concentrations. Between 1955 and 1985, an estimated 1.2×10^8 m^3 of waste rock and tailings were generated in the Elliot Lake and Bancroft areas. Assuming that the annual production rate of waste rock has remained constant, these tailings-management areas contain about 2×10^3 terabecquerels (TBq) of ^{226}Ra in 100 megatons of waste rock at a concentration of about 18.5 Bq/g (*8*). Radon emanates from these piles at a rate of about 22 Bq/m^2/sec (*9*). Estimates for total tailings production in the United States up to 1983 are on the order of 175 megatons (*10*).

Fuel Preparation. Preparation of fuel for nuclear power generation includes purification and conversion of the yellowcake concentrate to UO_3 or UF_6, isotopic enrichment in ^{235}U if required, and fabrication into reactor fuel elements. All fuel preparation facilities in the Great Lakes basin are located in Ontario, with various activities distributed between Blind River, Port Hope, Toronto, and Peterborough.

The principal waste generated by the conversion process is the raffinate from solvent extraction, although smaller quantities of uranium dust are released to the atmosphere. The main radioactive constituents are unrecovered ^{235}U, ^{232}Th, and ^{226}Ra, the latter which was discharged at a rate of about 48 GBq/year from the Port Hope refinery before its relocation to Blind River, Ontario, in 1983. This can be compared with the discharge of 90 GBq/day in the tailings of a single mill (*11*). Estimates of airborne uranium emissions from UF_6 production at Port Hope ranged from 252 kg in 1986 to 57 kg in 1989; quantities in liquid effluent ranged from 432 kg in 1986 to 65 kg in 1990 (*12*). Radionuclide releases from fuel fabrication facilities are small, with annual emissions to air from a 400 tons/year plant estimated to be about 0.52 GBq/year, equivalent to 3.5 kg/year of natural uranium (*11*).

Nuclear Power Generation. The first nuclear power reactor in the Great Lakes basin began operations in 1963 at Charlevoix, Michigan; in 1966, the first Canadian reactor at Douglas Point, Ontario, became operational. A 15-year period of growth in the nuclear industry was followed in the mid-1970s by a significant slowdown in the installation of additional nuclear capacity as a result of rising costs, environmental concern, and public pressure. There are presently 16 operational nuclear generating stations located on the shores of Lakes Michigan, Huron, Erie, and Ontario consisting of 36 pressurized and boiling water reactors (Table 3). Of the 27,000 MW of electrical generating capacity installed in the basin, just over 50% is generated in Canada.

Nuclear power generation results in the formation of artificial radioactive fission products within the fuel and the activation of stable elements in the structural material and coolant circuit. Although the fission product inventory is substantially larger than the inventory of activation products, the former is retained in the fuel core. Treatment systems remove the majority of radionuclides from the liquid and gaseous

Table 3. Commercial nuclear power plants in the Great Lakes basin.

Reactor name	Power, MW	Type	Startup year	Location
Lake Michigan—total installed capacity: 6400 MW				
Kewaunee	500	PWR	1974	Carlton, Wisconsin
Point Beach 1–2	2×485	PWR	1970–1972	Two Rivers, Wisconsin
Zion 1–2	2×1040	PWR	1973	Zion, Illinois
Donald C. Cook 1–2	2×1040	PWR	1975–1978	Bridgman, Michigan
Palisades	730	PWR	1971	South Haven, Michigan
Big Rock Point	67	BWR	1962	Charlevoix, Michigan
Lake Huron—total installed capacity: 7000 MW				
Bruce A	4×850	PHWR	1977–1978	Kincardine, Ontario
Bruce B	4×850	PHWR	1984–1987	Kincardine, Ontario
Douglas Point	200	PHWR	1966/1984[a]	Kincardine, Ontario
Lake Erie—total installed capacity: 3100 MW				
Fermi 1	Fast breeder, accident and shutdown 1966			
Fermi 2	1090	BWR	1986	Newport, Michigan
Davis–Besse 1	860	PWR	1977	Oak Harbor, Ohio
Perry 1	1140	BWR	1986	North Perry, Ohio
Lake Ontario—total installed capacity: 10,300 MW				
Pickering A	4×500	PHWR	1971–1973	Pickering, Ontario
Pickering B	4×500	PHWR	1982–1986	Pickering, Ontario
Darlington A	4×850	PHWR	1992	Newcastle, Ontario
Robert E. Ginna	470	PWR	1969	Ontario, New York
Nine Mile Point 1	610	BWR	1969	Scriba, New York
Nine Mile Point 2	1070	BWR	1987	Scriba, New York
James A., FitzPatrick	760	BWR	1975	Scriba, New York
Near basin—total installed capacity			8,900 MW	
12 additional reactors located in the near basin				
Great Lakes basin—total installed capacity			26,800 MW	
Great Lakes basin and near basin—total installed capacity			35,700 MW	

Abbreviations: MW, megawatt; PWR, pressurized water reactor; BWR, boiling water reactor; PHWR, pressurized heavy water reactor (CANDU). [a]Reactor shutdown date. Data from UNSCEAR (*4*).

Table 4. Radioactive wastes associated with the operation of the Pickering A (2000 MW) and Bruce A (3000 MW) generating stations for 1 year.

Stage in fuel cycle	Quantity	Principal radiation	Activity, TBq
Mining, milling, and refining (0.2% U ore)	1.6×10^5 tons	α	40.7
Fuel fabrication	200 m^3	α	0.015
Reactor operation			
Airborne effluent[a]	—	β	4800 (Pickering A+B) 1600 (Bruce A+B)
Liquid effluent[a]	—	β	600 (Pickering A+B) 750 (Bruce A+B)
Reactor wastes	1000 m^3	β,γ	74
Irradiated fuel	356 tons	α,β,γ	1.18×10^9 [b] 8.51×10^6 [c]

Based on 80% generating capacity. [a]Average 1985 to 1989 discharges (*4*); major component is tritium. [b]Activity at discharge. [c]Activity at one year after discharge. Data from UNSCEAR (*4*) and Atomic Energy of Canada Limited (*13*).

streams during purification. Low levels of radionuclides are released to the environment under controlled and monitored conditions, in quantities dependent on the reactor type and design. Atmospheric releases include tritium (^3H), radioiodine, fission product noble gases (^{88}Kr, ^{133}Xe), activation gases (^{14}C, ^{16}N, ^{35}S, ^{41}Ar), and particulates such as ^{60}Co, ^{90}Sr, and ^{137}Cs. Radionuclides released into the aquatic environment include ^3H and other fission products and activated corrosion products (*3,4*). Tritium in aqueous and gaseous emissions is the principal radionuclide released from Canadian CANDU (CANada Deuterium Uranium) heavy water reactors. As a condition of licensing, nuclear generating stations are required to monitor and report all releases to the responsible authority. The Atomic Energy Control Board (AECB) imposes a strict design objective for releases from CANDU power reactors of 0.05 mSv/year at the site boundary or 5% of the public dose limit. The U.S. Nuclear Regulatory Commission regulates doses at the boundaries of American reactor facilities.

Estimated cumulative inputs of ^3H, ^{90}Sr, and ^{137}Cs to the lakes from fallout and from liquid effluent releases from nuclear installations are given in Table 2 (*2*). A comparison of inventories shows that the contribution due to fallout is significantly greater than that from nuclear power installations. The relative contribution due to continued reactor operations can be expected to increase as the remaining fallout radionuclides decay.

Nuclear Fuel Waste Management. The final step in the nuclear fuel cycle is the management and disposal of radioactive wastes, some of which have extremely long half-lives. Wastes generated in the fuel cycle fall into the broad categories of high-level radioactive wastes, which comprise mainly unprocessed spent reactor fuel, and low-level radioactive wastes that comprise most other operational wastes. Whereas liquid and gaseous effluent may be released to the environment in a controlled manner, solid wastes are stored either at the facilities or in licensed waste consolidation areas. All spent fuel is currently stored at the reactor sites. Radioactive waste management options are determined by the type and origin of the waste and are subject to regulatory, technical, and sociopolitical considerations.

Nuclear power plant wastes consist of activation products in the coolant and structural materials, low-level solid and liquid wastes produced through reactor maintenance, and high-level spent reactor fuel and irradiated reactor components. Low-level wastes include in-reactor components, filter media, ion-exchange resins, contaminated clothing and tools, and laboratory wastes. All low-level wastes produced by Canadian reactors, or resulting from research and maintenance, are stored at the Bruce Nuclear Power Development on Lake Huron. Low-level wastes generated by American reactors are transferred to federally licensed sites for near-surface land disposal. Table 4 lists the quantities of radioactive waste associated with a single year of operation of a typical nuclear generating station (*4,13*).

By far, the largest quantity of radioactivity produced in the fuel cycle is contained within the irradiated fuel, which accounts for over 99% of the radioactivity produced during reactor operations. At discharge each fuel bundle contains approximately 10^7 to 10^8 GBq of radioactivity (*13*), most of which decays away within the first 1000

years, primarily due to the disintegration of ^{90}Sr, ^{137}Cs, and other fission products. Some actinides such as ^{238}Pu also decay significantly during this time (*14*). A typical 2000 MW capacity CANDU station produces about 350 tons of spent fuel each year as high-level waste (*13*). Spent reactor fuel is stored on site in water-filled containment pools with capacities for 5 to 10 years of irradiated fuel production. As the used fuel bays are filled, older spent fuel is moved to on-site dry storage containment.

The basic concept for the permanent disposal of high-level radioactive wastes is the containment and isolation of the material by burial in stable, underground formations. These formations would provide a natural barrier to the release of radioactivity, which would be further inhibited by solidification or vitrification of the waste before placement in the repository. The deep geologic waste disposal concept proposed by Atomic Energy of Canada Limited for Canadian high-level wastes is currently in the process of environmental assessment (*15*). Both the Canadian and Ontario governments have opted to delay site selection until after the approval of the disposal concept. Any future repository will likely be located in the Canadian Shield and possibly in the basin.

Incidental Sources and Low-level Wastes

The most significant source of radioactive wastes in the basin is the nuclear fuel cycle; however, many non-fuel cycle facilities, principally hospitals, universities, government laboratories, and industry, have been licensed to use radionuclides. The low activities and short half-lives of the radionuclides employed generally permit disposal through dilution and discharge into municipal sewer systems. Solid wastes are disposed of at low-level burial sites. Studies carried out to assess the relevance of these sources showed that the majority of radionuclides contained in sewer discharge were from natural or fallout origin (*16*). Medical and industrial discharges of radionuclides to municipal sewer systems from licensed facilities have little impact on the basin.

Certain technologies and industrial processes make naturally occurring radionuclides more accessible to the environment. The combustion of fossil fuels, such as coal for electric power generation, releases ^{238}U and ^{232}Th decay series radionuclides and ^{40}K in fly ash. Normal environmental levels of uranium and thorium are sufficiently high that changes due to emissions from coal-fired power plants are barely detectable (*10*). A study of emissions from a thermal generating station in southern Ontario revealed that atmospheric releases of radionuclides were insignificant compared to routine emissions from a nuclear generating station of similar capacity (*17*). Phosphate ores used in fertilizers may also release small quantities of radionuclides into the ecosystem. In general, however, the impact of incidental sources on the basin is negligible.

Areas of Local Contamination

Although the current levels of radionuclides in Great Lakes waters are below objectives specified in the Great Lakes Water Quality Agreement, some areas in the basin can be considered radiologically contaminated on a local scale. Contamination at these sites has resulted from nuclear fuel cycle or radionuclide operations.

Port Hope and Port Granby, Lake Ontario. Several sites in the Great Lakes basin have been contaminated as a result of early waste management practices. The most important example is the town of Port Hope, which lies approximately 100 km east of Toronto on the north shore of Lake Ontario. Before its current uranium conversion operations were opened, the town was the site of radium (1933–1953) and uranium (1953–1983) refining operations and fuel fabrication facilities. In 1983, uranium refining operations were relocated to Blind River, Ontario, and new facilities for UF_6 production were constructed. From 1933 to 1948, wastes from the radium operations were deposited at several sites within the town. These sites were replaced in 1948 by the Welcome Waste Management Facility in Hope Township. Disposal of waste at Welcome ceased in 1955 with the opening of the currently operating Port Granby Waste Management facility in the Town of Newcastle, first licensed by the AECB in 1976 (*8*). The refining of U_3O_8 concentrate from 1953 to 1983 generated about 25 TBq of ^{226}Ra, most of which was deposited at Port Granby (*2*).

Due to the absence of regulations, disposal of radium wastes in Port Hope before 1948 was not well controlled. Many of the waste sites were exposed at the surface, and significant quantities of these wastes were used as fill material for construction and landscaping activities. The main radioactive contaminants are uranium, ^{230}Th, and ^{226}Ra. Wastes that represented an immediate health hazard were removed from residential, commercial, and public buildings during remedial clean-up activities conducted by the AECB during the latter half of the 1970s. These wastes were consolidated at less accessible areas in the town.

Wastes at the Port Granby waste management facility, 16 km west of Port Hope, consist primarily of uranium, ^{230}Th, ^{226}Ra, and their decay products. The total estimated volume is approximately 348,000 m^3, with average activities of 0.1 GBq/m^3 of ^{230}Th and 0.07 GBq/m^3 of ^{226}Ra (*8*). Runoff and groundwater collected in reservoirs are pumped to a water treatment facility north of the burial site. Groundwater flowing from the site to the lake carries about 25 MBq of ^{226}Ra and 25 kg of uranium annually, both of which are diluted at the shore to concentrations that are below drinking water guideline levels (*2*).

The contamination of water and sediments in the Port Hope harbor due to the release of liquid wastes from the refinery has resulted in the designation of the harbor as an area of concern by the Great Lakes Water Quality Board. Concentrations of uranium and gross alpha–beta radioactivity in harbor waters are often above maximum acceptable values defined by the Great Lakes Water Quality Agreement (*18*). About 90,000 m^3 of sediment are contaminated with uranium and thorium decay series radionuclides, as well as other heavy metals. Typical contaminant concentrations in the harbor sediments are about 22 MBq/m^3 of ^{226}Ra and 310 μg/g of uranium (*8*). Though classified as low-level waste, it is unlikely that ^{226}Ra contamination of the harbor sediments has an effect on the human food chain [RW Durham, unpublished report; (*19*)]. Although a potential risk exists from direct contact with the sediments, the existing depth of water forms an effective barrier to exposure of the general population.

In addition to ore wastes, ongoing operations result in the routine release of uranium dust into the atmosphere. Monitoring studies have been conducted by Health Canada to estimate the impact of these emissions on human health. Estimated doses from inhaled dust resulting from 1 year of refinery operation were 0.044 mSv at the nearest monitoring station in 1988 to 1989 and 0.16 mSv in 1981 to 1982 (*20,21*). These doses, while below the guideline dose of 1 mSv/year recommended by the International Commission on Radiological Protection (ICRP) for public exposure (*22*), may contribute a significant fraction of the normal

background radiation of 2.6 mSv/year. No health effects would be discernible at these levels. Health Canada is currently in the process of reconstructing total cumulative doses to Port Hope residents resulting from all current and historical operations and waste management practices.

Serpent River Basin, Lake Huron. The Serpent River basin, located on the North Channel of Lake Huron and draining the Elliot Lake uranium mining region, has received elevated levels of natural radionuclides since the mid-1950s. Radiologic monitoring by the Ontario Ministries of Environment and Labour has shown increased concentrations of ^{226}Ra from mining activities. Remedial measures implemented in 1966 reversed this trend; however, until 1977, average annual concentrations of ^{226}Ra in the Serpent River exceeded both the Ontario Criterion for Public Surface Water Supply and the target concentration of the Canadian Drinking Water Quality Guidelines in effect at that time. In 1985, the Great Lakes Water Quality Objective for ^{226}Ra in water was met. Levels of radionuclides in sediments first measured in 1975 have been steadily declining as a result of improved waste management practices. Concentrations in sediments in 1984 were 40% of the 1975 levels. Continued monitoring of the watershed is carried out by provincial agencies (*2*).

West Valley Reprocessing Plant and Waste Disposal Facility, Lake Erie. The Western New York Nuclear Service Center at West Valley, New York, 65 km upstream of Lake Erie on the Cattaraugus Creek, was the first commercial nuclear fuel reprocessing facility in the United States. As a result of reprocessing activities, the site contains high-level liquid waste tanks, a high-level solid waste disposal area, and a spent fuel storage pool. In 1962, the site was licensed to receive low-level radioactive waste. These operations ceased in 1975 when water was found seeping from some of the low-level waste trenches, although controlled releases of radioactivity to the local watershed have continued. No spent fuel has been reprocessed since 1972. The facility is currently being used to investigate methods of encapsulation of on-site high-level waste as part of the West Valley Demonstration Project.

Although reprocessing and waste burial operations have ceased, the current inventory of wastes at the site present a long-term problem of disposal and contamination. Radioactive wastes stored at the site include about 2.32×10^6 liters of high-level liquid waste, 164 tons of uranium from spent fuel reprocessing, 3,900 m^3 of spent fuel components, and 66,000 m^3 of low-level waste (*9*). Studies on the local watershed have shown that Cattaraugus Creek water and sediment have been affected by releases from the facility. Radionuclides released during former reprocessing operations in 1969 to 1971 resulted in average ^{90}Sr levels that were above the U.S. Environmental Protection Agency's drinking water standards and the U.S. Nuclear Regulatory Commission's technical specifications for the facility. Radionuclides released from the low-level waste sites have been detected in Lakes Erie and Ontario. These radionuclides reside mainly in Lake Ontario sediment and indicate that any accidental release from the facility could be transported to the lakes (*2*).

Nuclear Emergencies

Although the probability of occurrence of a severe accident is small, the impact of a nuclear emergency must be considered in a radiologic assessment of the basin environment. It is unlikely that any catastrophic radiologic event would occur for the phases of the fuel cycle that deal with unirradiated fuel. Materials present before placement in the reactor are generally found in nature and have low specific radioactivity. A major accident at a conversion or fuel fabrication facility, while resulting in releases substantially larger than normal, would not yield a significant number of radiologic health effects to the area's population.

Estimation of both the probability and consequence of a severe accident during nuclear power generation is difficult. Estimates for some of the more common events, such as stuck control rods or loss of coolant, are in general relatively low in consequence insofar as individual exposures are concerned. For the less probable but more severe accidents that would result in an uncontrolled release of volatile radionuclides, the local consequences could be significant in terms of the health, social, or economic implications. However, risk assessments have shown that the probability of causing a given number of fatalities from a nuclear accident is orders of magnitude lower than other man-made or natural hazards (*10*).

A massive radionuclide release from a reactor beyond the basin could affect the Great Lakes ecosystem as a result of long-range atmospheric transport of airborne radioactivity. The impact of an accident would be dependent on the amount and type of material released, its chemical and biologic behavior in the environment, and the distance of the reactor from human populations. In the case of the 1986 Chernobyl accident, the small contribution to the gross beta activity in the basin is identifiable in Figure 2 only because the weapons fallout activity had decreased to levels that were no longer detectable. Similarly, ^{137}Cs from Chernobyl was identified in milk in 1986 and 1987 because the fallout from weapons testing was almost undetectable (Figure 3).

Transport, Behavior, and Distribution of Selected Radionuclides

The radiologic impact of a particular radionuclide in an ecosystem is a function of its environmental, biologic, and radiologic properties. Environmental availability and behavior are dependent on complex interactions between physical, chemical, and biologic parameters. Radioactive decay results in the depletion of the radionuclide in the environment or body. However, due to the general movement of radionuclides through the biosphere, the effective half-life of a radionuclide in a particular medium may be much less than its radioactive half-life.

The major pathways by which exposures to specific radionuclides occur are identified as critical pathways. Radionuclides that are in soluble form and chemically analogous to essential nutrient elements will tend to follow pathways similar to their nutrient analogues. As a result, they will be extensively and rapidly transferred through the food chain. For example, ^{90}Sr, ^{140}Ba, ^{226}Ra, and ^{45}Ca behave like calcium and are therefore bone-seeking elements; ^{129}I and ^{131}I behave like stable iodine and accumulate in the thyroid. Radionuclides distributed throughout the body include ^{40}K, ^{137}Cs, and ^{86}Rb (which follow the general movement of potassium), ^{3}H (which resembles stable hydrogen and is found as tritiated water), and ^{14}C (which is part of the carbon cycle.)

Water samples collected from the Great Lakes between 1973 and 1981 indicate a general decrease in radionuclide levels at a rate of 2 to 5% per year (*2*) due to radiologic and physical removal. The radionuclides of greatest concern in the basin, from a health perspective and in terms of the potential for normal or accidental release from nuclear fuel cycle facilities, are ^{3}H, ^{14}C, ^{90}Sr, ^{131}I, ^{137}Cs, and ^{226}Ra. Short descriptions of the more important radionuclides that may be found in the basin environment are given below.

Tritium. Tritium with a half-life of 12.3 years, exists in the environment mainly as water; from water it enters the hydrologic cycle and all components of the biosphere. It is produced naturally in the upper atmosphere and artificially in nuclear detonations and nuclear reactors. Nuclear weapons tests conducted in the atmosphere since 1945 have produced quantities of ^{3}H far exceeding the natural inventory. Tritium produced during nuclear reactor operation is released in liquid and gaseous effluent as tritiated water.

Exposure to environmental ^{3}H occurs primarily through the critical pathway of ingestion, with additional contributions from inhalation and absorption through the skin. Following ingestion, tritiated water is completely absorbed from the gastrointestinal tract and is rapidly distributed throughout the body via the blood. The majority of this amount is removed from the body with a biologic half-life ranging from 2.4 to 18 days, which represents the turnover of body water. The remainder is removed with a half-life of one month to one year, representing the turnover of ^{3}H incorporated in organic compounds (*23*).

Average ^{3}H concentrations in Canadian surface waters are approximately 5 to 10 Bq/L due primarily to residual fallout from pre-1963 weapons tests. Average levels in the Great Lakes ranged from 7 to 10 Bq/l during 1982 to 1984 (*24*) and from 9 to 11 Bq/l during 1991 to 1993 (*25*). Radioactivity concentrations in community water supplies near all Ontario nuclear reactors ranged from 12 to 35 Bq/l , which are slightly elevated with respect to background levels (*26*). By comparison, the proposed Canadian federal guideline for ^{3}H in drinking water has been set at 7000 Bq/l, based on an annual dose of 0.1 mSv and a water consumption rate of 2 l/day (27). Tritium is detectable in air in the vicinity of CANDU reactors although the levels are low. Concentrations decrease from about 2 to 3 Bq/m^3 at a distance of 3 km to background values of about 0.1 to 0.2 Bq/m^3 at 40 km (*6*). The yearly increase in ^{3}H levels in Lake Ontario due to routine CANDU operations has been projected to be about 0.12 Bq/l (*25*).

Carbon-14. In addition to natural production in the stratosphere and upper troposphere, ^{14}C is produced by nuclear weapons detonations and nuclear reactor operations. The ^{14}C injected by nuclear tests roughly doubled the natural steady-state radioactivity in the atmosphere (*28*). Production of ^{14}C in nuclear reactors varies with reactor type; ^{14}C released in liquid or gaseous effluent may be present as CO, CO_2, or CH_4.

Carbon-14 is of interest because of its long half-life (5730 years) and its availability in the environment. Once released, ^{14}C enters the global carbon cycle, eventually giving rise to increased levels in humans. Intake of carbon and subsequent exposure to ^{14}C is primarily through ingestion, with almost complete absorption by the body. Inhalation intake accounts for about 1% of total carbon intake of which very little is retained in the body. The mean residence time in the body is about 53 days (*23*).

Measurements of ^{14}C in the leaves of maple trees growing in Gatineau Park 20 km northwest of Ottawa, Ontario, reveal a smooth decrease in excess ^{14}C produced by nuclear detonations (*29*). These decreases are occurring at a rate much quicker than that based on radioactive decay alone, and by the turn of the century, ^{14}C levels will not be measurably elevated above natural levels (*7*). Since the main significance of ^{14}C results from its entry into the global carbon cycle, releases from nuclear reactors in the basin will give a more or less uniform radiation exposure to the world population over a number of generations.

Strontium-90. Strontium-90 has received extensive monitoring in the environment and in human food chains. It decays with a half-life of 29 years through ^{90}Y, which is also radioactive, to form stable ^{90}Zr. Strontium is metabolically similar to calcium, barium, and radium and follows calcium through the food chain from the environment to man. Both ^{90}Sr and calcium are retained largely in the bone. Strontium is produced by nuclear detonations and nuclear power generation. The majority of environmental ^{90}Sr has come from weapons fallout; discharge rates from nuclear reactors are very small and indistinguishable from fallout. The deposition of ^{90}Sr on land and the transfer to humans by ingestion of contaminated food is the most important exposure pathway. Significant transfer occurs via the air–vegetation–livestock–milk pathway. Of less importance are the aquatic pathways, and contributions from drinking water are always less than 5% of the total ingestion intake (*23*). Upon ingestion, absorption of ^{90}Sr by the body is relatively high. The mean residence time in bone tissue ranges from 3.4 to 6.7 years.

Mean activities of ^{90}Sr in the Great Lakes during 1981 to 1982 ranged from 15 mBq/l in Lake Superior to 29 mBq/l in Lake Ontario (*1*). Average concentrations recorded in the Winnipeg, Ottawa, and St. Lawrence Rivers and on Lakes Huron and Ontario near the Bruce and Pickering generating stations ranged from 1 to 12 mBq/l in 1988 (*6*). These levels are essentially all due to nuclear weapons fallout. The effective half-live for removal of ^{90}Sr from the lakes is of the same order of magnitude as its radioactive half-life, indicating that radioactive decay is the major mechanism in the removal of ^{90}Sr from the lake environment (*30*).

Radioiodine. As a volatile element, radioactive iodine has received extensive study in view of its mobility and its selective irradiation of the thyroid gland when taken into the body. It is found in the environment mainly as a result of nuclear explosions and nuclear reactor operation, although ^{129}I and ^{131}I are naturally present as a result of spontaneous fission of natural uranium. Of the 15 isotopes of iodine produced by fission in nuclear reactors, the ones of radiologic interest are ^{129}I ($t_{1/2}$ = 1.6×10^{7} years) and ^{131}I ($t_{1/2}$ = 8.04 days). Although ^{129}I is produced in significantly smaller amounts and is not identified in routine discharges from reactors, practically all of it is still present in the environment, whereas virtually all of the short-lived ^{131}I has decayed. Releases of ^{131}I from reactors are widely variable and depend on the reactor coolant leakage rate. Since it is a volatile element, ^{131}I is readily released to the atmosphere in the event of an accident.

On consumption, uptake of iodine by the blood from the gastrointestinal tract is complete and rapid. Iodine is an essential component of the thyroid hormone and, as a result, is selectively taken up and concentrated in the thyroid gland. The ICRP transfer model assumes that 30% of the iodine entering the blood is transferred to the thyroid; from there it is cleared with a half-life of about 120 days (*31*). The absorbed dose in the thyroid is about 1000-fold that in other organs and tissues.

The most significant exposure route for environmental radioiodine is the air–vegetation–livestock–milk pathway. Fresh milk dominates as the major source of ^{131}I intake in areas where milk is a major component of diet as a result of the large areas scavenged by cows and the short storage period of milk. The ^{131}I content of milk samples collected monthly from farms near nuclear generating stations in Ontario are usually less than the minimum detectable radioactivity of approximately 0.15 Bq/l (*26*). As a result of its short half-life, ^{131}I is only of concern immediately following a

significant release from a reactor. Releases of long-lived ^{129}I could potentially have an impact in terms of committed doses to present and future generations, but this effect is far less than that from ^{14}C, which is routinely monitored in the environment.

Cesium-137. Cesium-137 is one of the more important fission products due to its relatively high yield and its ability to bioconcentrate in some food chains. It has a radioactive half-life of 30.17 years and is produced in nuclear explosions with a $^{137}Cs/^{90}Sr$ ratio of 1.6. Cesium-137 is released during normal reactor operations primarily in aqueous effluent. As in the case of ^{90}Sr, weapons fallout over land is the most important source as far as committed doses to man are concerned. On land, it is strongly affixed to soil, which limits both its downward mobility and its availability for root uptake in plants. Fixation by sediments in aquatic environments is similar to soil and reduces its concentration in the water column. Direct atmospheric deposition is the primary mode of contamination for vegetation.

Cesium-137 metabolically resembles potassium and is bioconcentrated in a number of food chains including the air–vegetation–livestock chain, the air–lichen–caribou chain, and the freshwater-fish chain. The main contributions to dietary intake are grains, meat, and milk. Cesium-137 ingested by man is readily absorbed and becomes uniformly distributed in soft tissues, with minimal uptake by bone tissue. The biologic half-life of ^{137}Cs is a function of age and sex, with a representative retention rate of 110 days for 90% of the body burden (*23*).

Measurements of ^{137}Cs in milk samples around Bruce generating station on Lake Huron do not show levels elevated above the national average. Concentrations of ^{137}Cs measured in surface waters of the Great Lakes averaged about 0.5 mBq/l in 1992 (*6*). No enhancement in concentration was observed in the vicinity of reactor installations. Removal times of ^{137}Cs from the Great Lakes are less than 1 year, with a longer component of 5 to 20 years, suggesting re-entry into the water column from sediments (*30*). Great Lakes fish were found to contain concentrations of ^{137}Cs several thousand times higher than in ambient water (*2,30*).

Radium-226. Radium-226 occurs naturally in soils as a decay product of the ^{238}U series. It decays with a half-life of 1600 years to form ^{222}Rn. Uptake in food is the major pathway into the body. The contribution of drinking water to total intake is small when supplies are drawn from surface water, which typically display a narrow range of concentrations. Concentrations in groundwater sources, however, are highly variable and result mainly from the interaction between the groundwater aquifer and radium-bearing materials such as rock, soil, and ore deposits. When taken into the body, its metabolic behavior is similar to calcium, and an appreciable fraction is deposited in bone. The remainder is distributed more or less equally in soft tissues. Following the decay of ^{226}Ra to ^{222}Rn in bone, approximately 70% of ^{222}Rn diffuses to the blood and is exhaled (*3*).

Radium-226 concentrations in water samples measured at various sites across Canada between 1981 and 1984 ranged from about 1 to 13 mBq/l (*24*). Radium levels in water samples from Port Hope, Ontario, and Regina, Saskatchewan, averaged less than 5 mBq/l in 1988. Levels recorded during the same period in Elliot Lake, Ontario, ranged from 8 to 18 mBq/l (*6*). Radium levels measured in selected fruits and vegetables from Port Hope ranged from 0.04 to 40 Bq/kg (*32*). In general, higher levels of ^{226}Ra can be expected in areas containing uranium mining and milling operations or where rock containing high concentrations of the natural radionuclides is in contact with water.

Radon-222. Radon-222 is a chemically inert gas with a radioactive half-life of 3.82 days which is produced through the decay of ^{226}Ra. Its decay products form a series of short-lived radionuclides that decay within hours to ^{210}Pb ($t_{1/2}$ = 22 years). The principal dose is to the lung due to the inhalation and accumulation within the respiratory system of the short-lived decay products attached to inert dust normally present in the atmosphere. The radiation dose following inhalation constitutes the main portion of the natural radiation dose to man. Ra-226 in the earth's crust is the major source of ^{222}Rn. Although most of the radon produced in soil is retained in the earth where it decays, a small portion diffuses into the air. Indoor radon results from emanation of the gas from the soil under buildings and from water, building materials, and domestic gas. Enhanced levels are also found in the vicinity of uranium mine, mill, and tailings operations, and less importantly, in the vicinity of phosphate industrial operations.

A substantial body of literature exists on the risks associated with radon exposure. Miners of uranium and other minerals occupationally exposed to radon decay products are known to have been at increased risk for lung cancer under past conditions of exposure. Because radon gas is also naturally found in indoor air, questions about the potential lung cancer risks as a consequence on residential exposure have been raised. These studies have yielded conflicting results in terms of correlations between risk and residential exposures, which are typically much less than occupational exposures.

A rigorous case–control study on the risk of lung cancer from radon in indoor air was conducted in Winnipeg, Canada, between 1983 and 1990 (*33*). Winnipeg was chosen as a study site because it has indoor radon levels that are elevated with respect to other Canadian cities, including those in the Great Lakes basin. The study consisted of 738 individuals with confirmed primary lung cancer and 738 controls matched on age and sex. Radon dosimeters were place in all residences in which the study subjects had reported living in within the Winnipeg metropolitan area for at least 1 year. After adjusting for cigarette smoking and education, no increase in relative risk for any lung cancers was observed among the identified cases in relation to cumulative exposure to radon. Although there are areas within the basin in which radon concentrations are slightly higher than the mean concentrations in Canada and the United States (34 Bq/m^3 and 46 Bq/m^3, respectively), there are no regions of abnormally high concentrations that would lead to health implications (*4,34,35*).

Uranium. Uranium normally found in nature consists of three long-lived isotopes of mass numbers 234, 235, and 238, with half-lives of 2.45×10^5, 7.04×10^8, and 4.47×10^9 years, respectively. Uranium-238 accounts for 99.28% by weight of natural uranium and is usually in equilibrium with ^{234}U in soils. Uranium-238 and ^{235}U are, respectively, the parent radionuclides of the uranium and actinium radioactive decay series. Both ^{226}Ra and ^{222}Rn are part of the uranium decay series.

Uranium is both chemically and radiologicly toxic. In general, chemical damage to the kidneys following acute ingestion of natural uranium is more important than radiation damage; radiation injury becomes more important if exposure occurs as a result of chronic ingestion or inhalation. Inhaled uranium compounds may be retained in the lungs or transferred to other parts of the body where they become incorporated in bone tissue (*31*). Decay of the

uranium isotope and its decay products can result in cancers in these locations.

Uptake in food is the principal route of exposure, although uranium is one of the more important natural radionuclides that may be found in water supplies. In general, levels of uranium in both surface and ground waters are low, typically less than 1 µg/l; uranium concentrations in Lake Ontario averaged about 0.7 µg/l during 1981 to 1984 (*24*). However, substantially higher concentrations have been measured in both private and community groundwater sources across Canada (*6*). Elevated airborne concentrations exist only in the vicinity of uranium milling and refining operations; in this case, inhalation becomes a critical pathway of exposure. Concentrations of airborne uranium in Port Hope ranged from 0.02 to 76 ng/m^3 in 1988. Background values in southern Ontario are of the order of 0.1 ng/m^3 (*20*).

Risk Assessment in the Great Lakes Basin

Radiologic risk assessments, while containing many uncertainties and simplifying assumptions, nonetheless allow a comparison of the impact of human activities with the effect of natural background radiation. These assessments require an estimation of the risk of an attributable health effect as a function of dose. While the purpose of this paper is to determine the relative contribution of the various sources in the Great Lakes basin, a risk assessment based on internationally recognized radiation protection methodologies and risk coefficients has also been carried out. Because all readers may not agree with the choice of the risk coefficient used in this assessment, a brief discussion on the estimation of risk for low-dose exposures has been included. If the reader wishes to use a higher coefficient of risk than the one used in this assessment, then a greater number of attributable health effects will be predicted; however, the relative contribution of the various sources will not change.

Extensive epidemiologic analysis has been carried out on populations exposed to various levels of radiation dose. The main sources of epidemiologic data are the populations exposed to high doses of radiation, primarily the Japanese atomic bomb survivors. Estimates of risk for these groups have been derived by both the Committee on the Biological Effects of Atomic Radiations, known as the BEIR V Committee (*36*), and by UNSCEAR (*3*) based on lifetime risk projections calculated from ongoing epidemiologic studies. The lifetime risk estimates for fatal cancer following high dose and high-dose rate radiation are given as 8%/Sv in BEIR V and 11%/Sv in UNSCEAR.

Both organizations state that their risk estimates should be reduced for low dose exposures protracted over several months or years to account for a reduced effectiveness of the cell damage mechanism. Using a maximum reduction factor of 2, UNSCEAR (*4*) recommends a lifetime risk estimate of 5%/Sv for fatal cancer following a protracted whole-body exposure of low dose and low dose rate radiation. The ICRP (*22*), while relying mainly on the assessment of the Japanese survivors by organizations such as UNSCEAR and BEIR V, has taken into consideration the entire body of literature in their estimate of risk. The lifetime risk estimate for low-dose exposures as given in the 1990 recommendations of the ICRP is 5%/Sv for the entire population, based on a linear, no-dose threshold model. On the basis of copious and on-going research in human epidemiology, animal studies, and cell biology, these organizations conclude that the risk estimates at low doses are likely conservative.

Supplemental to the lifetime risk estimate for fatal cancer, the ICRP has also derived a risk for nonfatal cancers weighted for severity. In addition, an allowance has been made for hereditary disorders, although no direct evidence supporting these effects has been found in human offspring. The total risk coefficient for fatal and weighted nonfatal cancers, and hereditary effects, based on all epidemiologic data, has been estimated to be 7.3%/Sv. This risk coefficient accounts for all cancer types in the entire exposed population, including women and children. Although the risk of occurrence for some cancer types may be higher in either females (e.g., breast cancer) or males (e.g., prostate cancer), at present, the uncertainties involved in the estimation of risk preclude such specific lifetime risk factors.

The authors recognize that a number of recent studies have indicated higher risks to populations exposed to low doses than would be expected from the above risk coefficients (*37–40*). While some of these require further consideration, many are flawed by serious methodological errors. On the other hand, some studies have shown a possible beneficial or "hormetic" stimulating effect from small doses of radiation. The vast majority of epidemiologic studies, both past and present, have produced risk estimates consistent with BEIR V, UNSCEAR, and ICRP. For these reasons, the authors have chosen to use the 1990 ICRP recommendations of 7.3%/Sv for their estimate of risk because it represents a convergence of international scientific opinion.

Effects other than cancer, such as neurologic, developmental, and immunologic damage, have been observed only at extremely high doses of radiation and are generally assumed to be threshold effects. There is no substantial evidence to support the occurrence of health effects other than the increased risk of cancer in individuals exposed to low levels of radiation. These effects will therefore not be considered further.

Having established the coefficient of risk to be used in the assessment, the total, or collective, dose received by the exposed populations from the various radiation sources can now be estimated. The collective dose allows an assessment of detriment in terms of a predicted number of health effects that may occur in the total exposed population. It is obtained by multiplying the average effective dose by the number of people exposed. For example, an average effective dose of 2.6 mSv/year from natural background radiation will result in a collective dose of 9.36×10^4 man Sv/year to the basin population of 36 million. Once the collective dose has been evaluated, the number of health effects theoretically attributable to the exposure can be estimated using the derived lifetime risk coefficient.

Collective doses to local and regional populations from nuclear fuel cycle activities must be evaluated from environmental exposure models since the radioactivity concentrations resulting from fuel cycle effluent are very low in environmental samples. The environments receiving the modeled releases are chosen to represent broad averages containing typical features of existing sites. Based on worldwide emission data and population distributions representative of facilities sited in northern Europe and the northeastern United States, UNSCEAR (*4*) has obtained a collective dose for the global nuclear fuel cycle, normalized to total power output, of the order of 3 man Sv/GW/year. While indicative of the overall nuclear program, this normalized value is not representative of any one site; in fact, large variations may be expected between different reactor types or management practices. To provide a more realistic assessment of the effect of fuel cycle activities, collective doses have been calculated based on

measured releases from basin facilities between 1985 and 1989 (*4*) and on a population density more representative of the basin, which includes the sparsely populated areas of northern Ontario.

Using UNSCEAR model values for collective doses per unit release of specific radionuclides from typical facilities, doses in the basin can be estimated from measured emission data. Collective doses from mining and milling facilities are based on radon releases from uranium mines in the Elliot Lake area; doses from reactor operations are derived from the measured emissions for all reactors in the Great Lakes basin (*4*). Doses for conversion facilities are minor and are therefore taken directly from UNSCEAR values normalized to global energy production. A factor of 0.5 was applied to all collective doses to account for the smaller population density around the basin as compared to that used in the UNSCEAR models.

In addition to the collective dose for the entire exposed population, the impact of anthropogenic sources is also assessed in terms of the dose to the maximally exposed or critical group. The critical group dose provides an indication of the greatest detriment that may occur to those individuals who live near a facility, and who derive all their food and water from local supplies.

Estimates of critical group and collective doses for the various natural and anthropogenic sources are shown in Table 5. The greatest contribution to total exposure, in terms of both individual and collective dose, comes from natural background radiation. Of significantly less importance is the dose resulting from nuclear weapons fallout. The average fallout dose to an individual is based on an incomplete dose to the year 2050 for radionuclide deposition in the North Temperate Zone, which provides a measure of the total radiation hazard from fallout presented to those living during the period of intensive atmospheric weapons testing before 1963. It is expressed as an integrated dose rather than an annual dose rate since most of the exposure from weapons fallout has already been received, and the dose from several other fallout radionuclides will have largely been completed by the end of the decade.

Estimated doses to individuals in the critical group from fuel cycle operations occur in the vicinity of operating mines and are approximately 0.6 mSv/year (*14*); although these estimated doses are below the ICRP dose limit of 1 mSv/year, they are nevertheless a significant fraction of this limit. The maximum allowable dose to critical groups in the vicinity of nuclear reactors is 0.05 mSv/year based on the design objective imposed by the AECB for public exposure at the boundary of Canadian nuclear generating stations. Actual estimated doses to the most exposed individuals in the vicinity of CANDU reactors based on Ontario Hydro monitoring programs were in the range of 0.01 to 0.04 mSv/year between 1991 and 1993 (*41*). Collective doses derived from measured emissions are several orders of magnitude less than that from natural background radiation and are due mainly to radon releases from mining activities and tritium and ^{14}C discharged by heavy water reactors.

Also shown in Table 5 are estimates of collective dose and risk committed by 50 years of exposure centred on the present. This standardization allows the risks from all sources to be expressed on a common basis, since not all doses are given as an annual rate. It is assumed that current annual values for natural background radiation and nuclear fuel cycle exposure are representative of this period. Based on ICRP risk coefficients and assuming a conservative, linear, no dose-threshold model, theoretical limits of risk can be estimated for the various radiation exposures in the basin over this period. The total number of theoretical fatal cancers, nonfatal weighted cancers, and hereditary disorders over the lifetime of the current basin population theoretically attributable to a 50-year exposure to natural background radiation is approximately of 3.4×10^5. By comparison, the total number of theoretical health effects attributable to radioactive fallout from all weapons tests to date would be approximately 5.0×10^3. Theoretically, attributable health effects due to 50 years of operation of the nuclear fuel cycle at current levels would be approximately 2×10^2. These numbers are hypothetical values based on conservative exposure models, rather than predictions of actual effects from either natural or artificial sources. However, they show that the impact in the basin from man-made sources are very small compared to the effects of normal background radiation.

The risks associated with a severe nuclear emergency are more difficult to predict. Although the probability of a severe accident has been shown to be extremely small, serious health effects could occur near the accident site if a massive release of radioactivity resulted from a breach in the reactor containment. Long-range transport and dispersion of the radioactive plume could result in the exposure of many people to marginally or significantly elevated levels of radiation. Additional future deaths due to cancer could occur as a result of increased collective doses. However, due to the engineered safeguards in North American reactors, it is expected that the social and economic consequences of an accident would predominate over actual health effects.

Concentrations of important radionuclides in Great Lakes waters that would result in a 50-year committed effective dose equal to the proposed Canadian federal drinking water guideline of 0.1 mSv from a single year's consumption of drinking water (730 l) are shown in Table 6 (*27*). These are compared with actual measured concentrations, which are well below the derived

Table 5. Maximum individual and collective doses and risks from radiation sources in the Great Lakes basin.

Source	Annual dose		50-Year collective dose and risk[a]	
	Individual[b], mSv/year	Collective, man Sv/year	Collective dose, man Sv	Risk[c] (health effects)
Natural	2.6	94,000	4.7×10^6	3.4×10^5
Fallout	1.9[d]		6.8×10^4	5.0×10^3
Nuclear fuel cycle				
Mining, milling[e]	0.65	15		
Conversion[f]	0.044	0.1		
Reactor operation[g]	0.01–0.04	40		
Low-level waste		<0.1		
Total fuel cycle		55	2.8×10^3	2×10^2

[a]Collective dose and risk for natural and fuel cycle exposures are integrated over 50 years based on current dose rates and integrated to the year 2050 for weapons fallout exposures; collective doses for nuclear fuel cycle are based on dose per unit release and measured emissions for basin facilities from 1985 to 1989. [b]Individual dose is the average population dose for natural and fallout exposure; for nuclear cycle exposures, it is the dose to individuals of the critical group. [c]Risk is defined in terms of theoretically attributable fatal and weighted nonfatal cancers and hereditary disorders, using the ICRP risk coefficient of 7.3%/Sv. [d]mSv to 2050. [e]Maximum dose for mining activities form NCRP estimates; from NCRP (*14*). [f]Maximum dose for conversion from Health Canada estimates; from Ahier and Tracy (*20*). [g]Maximum dose for the reactors based on Ontario Hydro estimates; from Ontario Hydro (*41*).

Table 6. Comparison of proposed Canadian federal guideline concentrations for radionuclides in water and actual concentrations in the Great Lakes.

Radionuclide	Guideline concentration, Bq/l	Observed concentration, Bq/l				
		Superior	Michigan	Huron	Erie	Ontario
^{3}H	7000	5.4	6.6	9.1	12	8.7
^{90}Sr	5	1.5×10^{-2}	1.9×10^{-2}	2.7×10^{-2}	2.3×10^{-2}	2.9×10^{-2}
^{137}Cs	10	1.7×10^{-3}	1.4×10^{-3}	1.1×10^{-3}	0.6×10^{-3}	1.0×10^{-3}
^{226}Ra	0.6	—	—	0.7×10^{-3}	—	1.2×10^{-3}
$^{239,240}Pu$	0.2	—	4.4×10^{-7}	4.8×10^{-7}	1.8×10^{-7}	1.7×10^{-7}
U, μg/l[a]	150	0.08	0.38	0.39	0.59	0.42

Maximum allowable concentrations in water are based on an annual exposure limit of 0.1 mSv and an annual water consumption of 730 liters. Water concentrations from International Joint Commission (*1*) and Joshi (*2*).
[a]Uranium concentration given in micrograms per liter; the guideline concentration corresponds to approximately 4 Bq/l and the limit based on chemical toxicity is 100 μg/l.

Table 7. The 50-year committed effective dose from the ingestion of Great Lakes water for 1 year.

Radionuclide	50-Year committed effective dose, μSv				
	Superior	Michigan	Huron	Erie	Ontario
^{3}H	0.08	0.09	0.1	0.2	0.1
^{90}Sr	0.4	0.5	0.7	0.6	0.7
^{137}Cs	0.02	0.02	0.02	0.01	0.01
^{226}Ra	0.2	0.2	0.2	0.2	0.2
U (natural)	0.04	0.2	0.2	0.3	0.2
Total dose, μSv	0.7	1.0	1.2	1.3	1.2

Average risk is three theoretically attributable health effects per year. The dose is based on concentrations from Table 6 except for ^{226}Ra for which a concentration of 1 mBq/l is assumed. The average basin risk is based on a committed effective dose of 1.2 μSv for a population of 36 million.

maximum concentrations. The effective doses from drinking water for each lake are shown in Table 7. The total average dose received from drinking Great Lakes water is estimated to be about 1.2 μSv for Lakes Ontario, Erie, and Huron and 1.0 μSv for Lake Michigan. These are well below the ICRP exposure limit and would result in three theoretically attributable fatal and weighted nonfatal cancers and hereditary disorders per year based on the maximum effective dose to the entire basin population. As with other estimates of risk, this estimate is an upper limit based on the conservative assumption of a no-threshold dose model.

Conclusions

The human population within the Great Lakes basin is continuously exposed to ionizing radiation in the environment from both natural and anthropogenic sources. The greatest contribution to total exposure is the natural background radiation that originates from both cosmic and terrestrial sources and results in an average dose of about 2.6 mSv/year to every resident of the basin. Global fallout from weapons tests has resulted in the largest input of anthropogenic radioactivity into the lakes, although the moratorium on atmospheric detonations has resulted in declining levels since the mid-1960s. The total committed dose to the year 2050 to an average individual in the basin from all weapons tests has been estimated to be about 1.9 mSv. The small but routine input from the large number of facilities comprising the nuclear fuel cycle is of increasing importance. Almost every stage of the fuel cycle is active in the basin, including mining, conversion, power generation, and waste management. Although the potential exists for a serious accident resulting from the large inventories of radionuclides contained in the reactor core and spent fuel bays, the probability of a such an occurrence is extremely small because of strict design and operational regulations. Serious accidents outside of the basin may also impact on local ecosystems as a result of long-range atmospheric transport of radioactive plumes. An area of importance over the next few decades will be the management of the substantial amounts of high-level wastes generated by the many reactors in the basin.

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