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IMPACT OF CANDU EMISSIONS ON TRITIUM LEVELS IN THE GREAT LAKES

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ABSTRACT

A concentration-time model has been developed to estimate the relative contributions of cosmogenic, weapons fallout and CANDU tritium emissions from OPG Nuclear Generating Stations to the Great Lakes tritium inventories. Tritium input to the lakes is from tritium in precipitation, water vapour exchange, drainage from lake watersheds and direct input from CANDU reactors. Removal is by outflow from the lakes, evaporation and radioactive decay. Lakes Huron and Ontario are the only two Great Lakes which receive direct inputs from CANDU reactors and are therefore of particular interest. Estimated relative contributions of cosmogenic, residual weapons testing fallout and CANDU tritium to 1999 lake tritium inventories are 6, 30 and 64% for Lake Huron and 8, 27 and 65% for Lake Ontario. Projections are that the contribution of CANDU tritium to Lakes Huron and Ontario tritium inventories will increase to 80% and the residual tritium fallout from historic weapons testing will decline to 10% by 2025. Lakes Huron and Ontario tritium concentrations are currently approximately 7 Bq/L; three orders of magnitude lower than the Ontario drinking water standard. These concentrations are projected to decline to 5 Bq/L by 2025. The decline is primarily due to the continued decay and outflow of residual weapons fallout tritium.

INTRODUCTION

Tritium in the hydrologic cycle is in the form of HTO and is derived from three sources: i) cosmogenic, ii) nuclear weapons fallout, and iii) emissions from industry. Modern day tritium levels of all five Great Lakes are dominated by weapons fallout from nuclear weapons tests performed in the atmosphere between 1954 and 1963 (Joshi, 1991). These resulted in a two order of magnitude increase in lake tritium concentrations. Since the 1970's, tritium emissions from CANDU reactors at Lakes Huron and Ontario have

impacted the tritium levels in these lakes and Lake Erie which receives inflow from Lake Huron.

The relative contribution of CANDU emissions to lake tritium concentrations will increase as residual tritium from historic weapons testing fallout continues to be removed from the lakes by outflow and decay. A preliminary assessment of the contribution of CANDU tritium to lake concentrations was performed by King and Workman in 1998 (King, written communication, 1998). This assessment concluded that CANDU emissions account for 50 and 65%, respectively, of the current Lake Huron and Ontario tritium inventories. The remainder is from cosmogenic and residual weapons fallout.

At the request of Ontario Power Generation (OPG) a concentration-time model was developed to more accurately define the inventories of cosmogenic, weapons fallout and CANDU-derived tritium, in the Great Lakes. This paper describes:

i) the theory and implementation of the model,
ii) cosmogenic, weapons fallout and CANDU tritium inputs to the Great Lakes, and
iii) model predictions of the relative contributions of cosmogenic, weapons fallout and CANDU tritium emissions to lake concentrations.

The model is evaluated using measurements of lake concentrations dating back to the early 1950's and, from this, projections of future tritium concentrations to 2025 are made.

THEORY

The Great Lakes can be visualized as a chain of well mixed lakes; Lakes Superior and Michigan are at the top of the chain and discharge to Lake Huron; Lake Huron discharges to Lake Erie which in turn discharges to Lake Ontario. Each lake receives inflow from the lake watersheds, defined as the land area draining to the lake, upstream lakes where applicable and direct precipitation. Removal of water is by outflow to downstream lakes and evaporation. There is some seasonal and annual variability in lake volumes, inflows and outflows. However, as lake concentrations for the period of interest span 75 years, 1950 to 2025, the effect of these variations on model predictions are considered minimal. The lakes water budget is assumed to be at steady-state.

The concentration-time model is based on a tritium mass balance for each of the Great Lakes. It is similar to the computational models reported in Lerman (1972) and Milton et al. (1994, 1997) which were used to evaluate the impact of weapons fallout ^{90}Sr and ^{36}Cl on the concentrations of these nuclides in the Great Lakes.

Tritium input to the lakes is from tritium in precipitation, water vapour exchange between the lake surface and atmosphere (Weiss and Roether 1979; Simpson 1970), inflow from lake watersheds and CANDU emissions. Tritium is removed by outflow, evaporation and radioactive decay. Performance of a tritium mass balance for each of the Great Lakes

yields a differential equation giving the change in lake concentration as a function of time (Lerman, 1972) . The differential equation is written as

$$\frac{dC_m}{dt} = \frac{W_m(t)}{V_m} + \frac{C_{m-1}q_{m-1}}{V_m} - \alpha_m C_m \qquad (1)$$

where the subscript *m* is the lake number in the chain, *C* is the lake tritium concentration, *t* is time, *W(t)* is the tritium input from precipitation, water vapour exchange between the lake surface and atmosphere, watershed inflows and CANDU emissions, *V* is the lake volume, *q* is the inflow from the upstream lake and α is a constant which accounts for removal of tritium from the lakes by radioactive decay and outflow. The second term on the right side of Equation 1 represents tritium inflow from upstream lakes and is equal to zero for Lakes Superior and Michigan as neither receives any significant amount of inflow from upstream lakes.

The sum of tritium inputs to each lake, *W(t)*, is written as:

$$W(t) = PC_p + E\frac{1}{fr}\left(\frac{h}{1-h}\right)C_p - E\left(\frac{1}{fr(1-h)}\right)C + C_D D + R \qquad (2)$$

where the first term describes tritium input by direct precipitation to the lake surface, the second and third terms represent net tritium input by water vapour exchange, the fourth term represents tritium input through inflow from lake watersheds and the fifth term *R* represents tritium input from CANDU emissions.

Tritium input by direct precipitation is the product of the precipitation *P* and tritium concentration in precipitation, C_p .

The net tritium input from vapour exchange is calculated according to Weiss and Roether (1979). This pathway is important for large water bodies and can exceed the tritium input through direct precipitation (Weiss and Roether, 1979; Simpson, 1970). Input by vapour exchange is proportional to the evaporation rate *E* and depends on the water vapour gradient in the atmospheric boundary layer (*h*: water vapour content at 10 m above sea surface-level relative to saturation; *h*=1 at the air-water interface). The "ingoing" fraction is set proportional to the tritium concentration of precipitation. The assumption is based on observational evidence that the tritium concentration in marine water vapour at ships height has a tritium concentration corresponding to isotopic equilibrium with falling precipitation (Weiss and Roether, 1979). The "outgoing" fraction is set proportional to the tritium concentration of the lake; *fr* is the isotopic fractionation factor.

Tritium input from the lake watersheds is the product of the tritium concentration of the watershed inflow C_D and the inflow rate *D*. The flow path of tritium deposited in the watershed to the lakes is not well characterized. Some of the deposited tritium is carried away by surface runoff and enters the lakes in the year of deposition, the remainder enters

groundwater where it is held up. Milton et al. (1997) describe the delay in groundwater using the generalized Poisson distribution written as:

$$f(T) = kT^n \exp(-T / \tau) \quad (3)$$

where k is a normalizing constant given by $\tau^{n+1}/n!$ and T is the groundwater residence time (Milton et al., 1997). The distribution $f(T)$ has a maximum at time $n\tau$ and zero slope at $T = 0$ except when n=0 or 1. The value of $n\tau$ approximates the mean groundwater residence time for the watershed. The groundwater residence times of the watersheds are not well characterized. The sensitivity of model predictions to the parameter values n and τ was evaluated and is discussed in the section **Sensitivity Analysis**.

The tritium concentration of the watershed inflow to the lakes is given by:

$$C_D(t) = (rf)(C_p(t)) + (1-rf)\int C_p(t-T) f(T) \exp(-\lambda T) dT \quad (4)$$

where rf, the runoff fraction, is the fraction of precipitation to the basin which runs off as surface water in the year of deposition, $(1-rf)$ is the fraction of precipitation to the basin which enters groundwater and λ is the decay constant for tritium. Milton et al. (1997) assumed a runoff fraction of 30% and this value is used here. It is comparable to the value of 45% derived from a study of a small watershed to the north of Lake Ontario (Howard et al., 1993).

The rate of tritium removal from the lakes is given by:

$$\alpha = \lambda + \frac{q}{V} \quad (5)$$

where the first term is the radioactive decay constant and the second term is the rate constant for removal by outflow; q is the lake outflow rate and V is the lake volume. Removal of tritium from lakes by evaporation is included in the expression describing net input by vapour exchange, Equation 2, and is not included in Equation 5.

Equation 1 is solved numerically using a fourth order Runga Kutta method. The time step for the calculations is 1 year. The solution method with time step 1 year is not suitable for Lake St. Clair, a small lake with residence time of less than 1 week, lying between Lakes Huron and Erie. The Lake St. Clair tritium concentrations are assumed equal to Lake Huron tritium concentrations.

DATA

Hydrologic Data

The complete Great Lakes drainage basin has an area of 791900 km^2 (246338 km^2 of water surface and 545594 km^2 land drainage). Great Lakes and Lake St. Clair hydrologic data (for example lake volumes, inflows from precipitation and lake watersheds) are listed in Table 1. The lake watershed is the area of land draining to the lakes. The data are from the Canadian Hydrological Atlas (various dates) and were compiled by Milton et al. (1994).

Outflow is an important removal mechanism of tritium for the lower lakes (Huron, Erie and Ontario). Concentration half-lives accounting for both outflow and decay for these lakes are 6.7 years for Lake Huron, 1.5 years for Lake Erie and 3.5 years for Lake Ontario, much less than the half-life for decay alone (12.34 years). Lake Superior and Michigan have much longer water residence times and tritium removal from them is governed by decay. Concentration half-lives due to both outflow and decay are 11.2 years for Lake Superior and 10.7 years for Lake Michigan.

Table 1 Hydrologic Data for the Great Lakes

	Superior	Michigan	Huron	St. Clair	Erie	Ontario
Area [km^2]	82367	58016	60536	1190	25220	19009
Volume [km^3]	12221	4586	3682	3.6	484	1635
Outflow [m^3/s]	2255	1275	5507	5609	6370	7374
Residence time [a]	171.88	114.08	21.2	0.02	2.41	7.06
Watershed area [km^2]	138586	117408	133886	15879	64563	75272
Watershed area/Lake area	1.68	2.02	2.21	13.34	2.56	3.96
Inflow precipitation [$m^3/a \times 10^{-9}$]	60.7	45.7	47.7	1	21.8	16.4
Inflow watershed [$m^3/a \times 10^{-9}$]	52.5	34.4	47.6	3.2	22.1	28.9
Inflow upper lakes [$m^3/a \times 10^{-9}$]			111.3	173.7	176.9	200.9
Total inflow [$m^3/a \times 10^{-9}$]	113.2	80.1	206.6	177.9	220.8	246.2
Outflow rivers [$m^3/a \times 10^{-9}$]	71.1	40.2	173.7	176.9	200.9	231.7
Outflow evaporation [$m^3/a \times 10^{-9}$]	42.1	39.9	30.9	1	19.9	14.5
Evaporation/volume [%]	0.34	0.87	0.84	38.55	4.11	0.89

Tritium Data

Cosmogenic and Weapons Fallout Tritium

The most significant natural source of tritium in the environment is from the interaction of neutrons generated by cosmic rays with N_2 and O_2 of the upper atmosphere, (Stewart and Farnsworth, 1968). An equilibrium quantity of 3.5 kg of tritium is present from these sources (Michel, 1976). Tritium in the atmosphere is oxidized to HTO and deposited to the earth's surface by rain and water vapour exchange. The concentration of cosmogenic tritium in precipitation has been estimated to be 0.24 -1.18 Bq/L depending on geographical location and weather phenomena (Kaufman and Libby, 1954). We used the upper limit to estimate the cosmogenic tritium input to the Great Lakes.

The major source of tritium now present on the earth has been from the detonation of nuclear bombs (Michel, 1976). Detonation of the first uranium and plutonium fission bombs in the atmosphere began in 1945. The neutron flux from these comparably small fission tests had no significant impact on tritium levels in the environment. Major releases of bomb tritium occurred in 1954, 1956, 1958 and particularly in 1961 and 1962. In 1963 a partial test ban treaty came into force. The amount of tritium released from nuclear weapons tests since then has been a minor addition to existing levels (Brown, 1989). Michel (1976) estimated the total tritium produced by nuclear explosions to have been 550±160 kg, about 150 times the natural global inventory.

The rise in tritium concentration in precipitation due to weapons testing has been monitored worldwide. The International Atomic Energy/World Meteorological Organization (IAEA/WMO) database (1998) provides records of monthly average tritium concentrations from selected monitoring stations. These records represent cosmogenic and weapons fallout tritium. Records are provided for three monitoring stations in the vicinity of the Great Lakes Basin: Ottawa Ontario (45.32 N 75.67 W) located 160 km northeast of Lake Ontario; Madison Wisconsin (43.13 N 89.32W) located 125 km west of Lake Michigan; and Chicago Illinois (41.78 N 87.75 W) lying on the southwest border of Lake Michigan. Only the Ottawa record provides continuous monthly data for the period preceding major bomb releases to present (1953 to 1992) and we used it to estimate the weapons fallout source term for the Great Lakes.

Figure 1 shows 12 month average tritium concentrations in Ottawa precipitation for the period 1954 to 1992. The plot shows the rise in tritium concentrations following the major weapons releases and the subsequent decline following the partial weapons testing ban in 1963. The major tritium peaks in precipitation follow atmospheric nuclear weapons tests by 1 to 2 years. Tritium concentrations peaked in 1965 at which time they were 2 orders of magnitude greater than cosmogenic. Since 1980 tritium levels have remained relatively constant.

Tritium concentration in precipitation is variable over the earth's surface (Weiss and Roether, 1979; Brown 1989). Over North America, the general pattern is higher

concentrations in mid continental regions than in coastal regions and increasing concentration with increasing latitude (Stewart and Farnsworth, 1968). Some spatial variability in tritium concentrations in precipitation over the Great Lakes is observed. Monthly average tritium concentrations at the northern most station in the basin (Ottawa) are a factor 1.2 greater than at the southernmost station (Chicago). This suggests that use of the Ottawa tritium record could result in an over-prediction of tritium input to the basin by 20%. These latitudinal effects were evaluated in a sensitivity analysis and the tritium inputs were adjusted accordingly (Section **Sensitivity Analysis**).

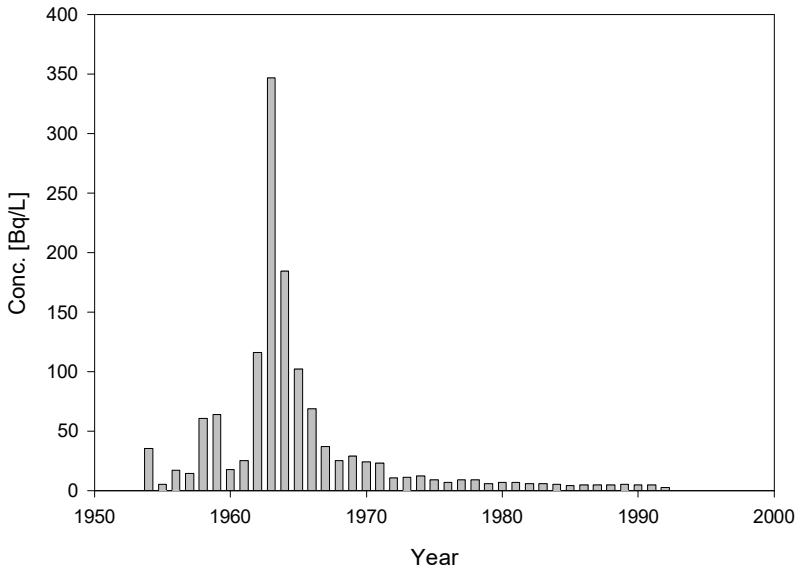

Figure 1 Ottawa tritium precipitation record, IAEA/WMO database (1998); Twelve month average data, 1954 -1992.

CANDU Tritium Emissions

Liquid and airborne HTO emissions from the 8 CANDU reactors in operation on the shores of Lake Huron and 12 on the shores of Lake Ontario are the primary source of CANDU HTO entering the Great Lakes. An additional source is the tritium removal facility at Darlington on the shore of Lake Ontario which releases HT to the atmosphere. However, only a small amount is expected to oxidize to HTO (Davis, personal communications, 1999). The total HTO contribution of the Darlington tritium removal facility to annual emissions is less than a few percent.

Annual liquid and airborne tritium emissions at Lakes Huron and Ontario since the start up of the first CANDU reactors on Lake Huron in 1969 and Lake Ontario in 1971 are

shown in Figure 2. The data are from John LaMarre (personal communication, 1999), Gorman and Wong, (1978) and AECB (1987).

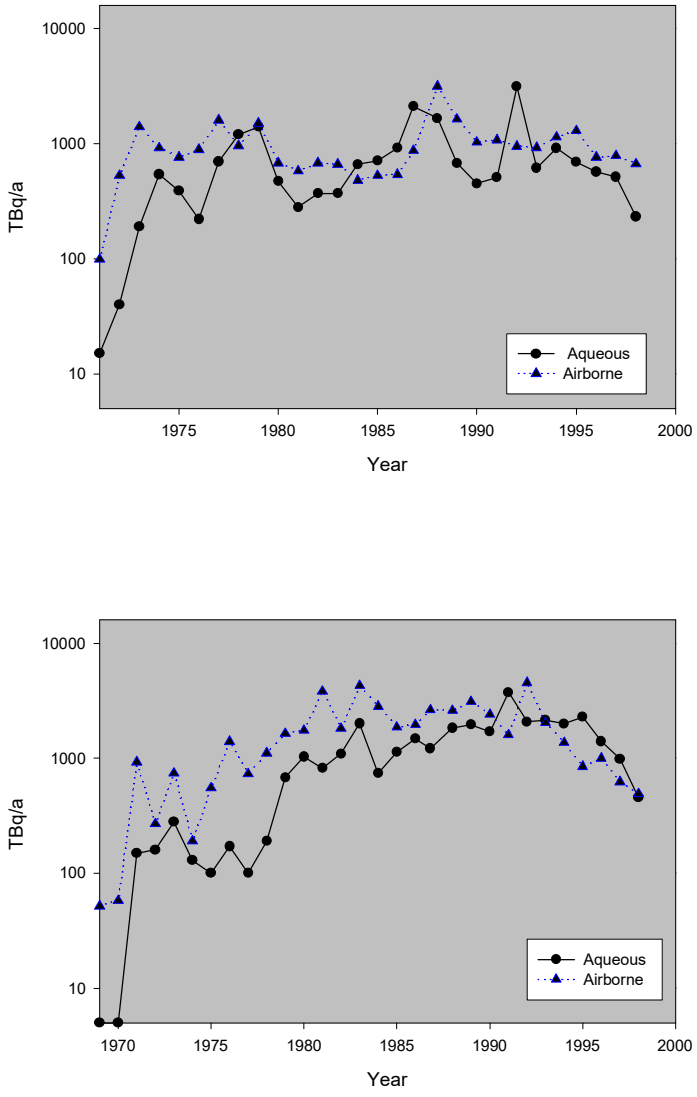

Figure 2 CANDU tritium Emissions at Lake Ontario (top) and Lake Huron (bottom).

Liquid tritium emissions to Lake Ontario have ranged from 5 to 3737 TBq/a with an average of 1068 TBq/a over the period between 1971 and 1998. Liquid tritium emissions to Lake Huron ranged from 15 to 14000 TBq/a with average of 1570 TBq/a. Airborne tritium emissions are of the same order of magnitude as the liquid emissions.

Tritium from airborne releases enters the lakes by deposition to the lake surface and by deposition to lake watersheds and subsequent runoff. The fraction of airborne released tritium entering the lakes is not known (Joshi 1991). Gorman and Wong (1978) assumed

50% of airborne tritium enters the lakes in their radiological assessment of airborne tritium emissions. This estimate is considered to be conservative (P.A. Davis, personal communications, 1999) and we have used it in our analysis.

Great Lake Tritium Concentrations - Measured Data

Measured Great Lake tritium concentration data for the period between 1953 and 1997 are used to evaluate the model. Table 2 lists measured tritium concentration data which were compiled from several sources and provided by King (K. King, written communications, 1998, 1999).

Table 2 Measured Great Lake Tritium Lake Concentrations: 1953 to 1997

Superior		Michigan		Huron		Erie		Ontario	
Year	Bq/L	Year	Bq/L	Year	Bq/L	Year	Bq/L	Year	Bq/L
1966	11	[e]1953	0.19	1966	21.5	1958	11.5	1958	8.1
1969	13	1960	3.4	1969	21	1966	36.5	1965	43
[a]1973	11	1965	19.2	[b]1974	14.2	1969	27.3	1966	26.7
[a]1981	6.7	[a]1981	7.4	[a]1981	10.6	[a]1973	12.6	[a]1973	11.1
1982	5.4	[f]1998	3.0	1982	9.1	[b]1974	15.3	[b]1974	16.4
[d]1997	2.0			[c]1991	7.2	[a]1981	8.5	[a]1981	13.5
				[d]1997	7	[c]1991	6.6	[c]1991	9
						[d]1997	5.5	[d]1997	7.1

a open water samples, (Joshi, 1991)
b sampling from at least 2 stations per lake and over depth of water column, (Torgerson et al. 1977)
c whole lake surveys for Lakes Erie and Ontario and survey of Detroit River outflow from Lake Huron (King, 1997)
d Superior, Huron, Erie and Ontario whole lake surveys (King, 1998, written communication)
e samples taken at Chicago (Libby, 1955)
f Lake Michigan whole lake survey (King, 1999, written communication)

The data show the rise in tritium concentrations to the mid 1960's and the subsequent decline owing to decreasing weapons fallout input and removal of tritium from the lakes by decay and outflow.

RESULTS AND DISCUSSION

The sensitivity of the model predictions to uncertainties in groundwater residence time of the lake watersheds and the cosmogenic and weapons fallout source term was first evaluated. The model was calibrated using test data for the pre-CANDU period, prior to 1970. Lakes Superior and Michigan are not affected by CANDU releases and all data for these lakes were used. The calibrated model was then used to compute the contributions of the cosmogenic, weapons fallout and CANDU source terms to the total lake concentrations and predict these concentrations to 2025.

Sensitivity Analysis

Watershed residence time: The delay of tritium in the watershed groundwaters is not precisely known. It is probably of the order of 5 to 10 years. In addition to the lag in tritium input to the lakes from the watersheds, the decay associated with the delay reduces input to the lakes. Predictions for: i) a mean residence time in lake watersheds of 9 years ($n=3, \tau=3$, Equation 3), and ii) no delay in lake watersheds are shown for Lake Huron (Figure 3). For all lakes, the hold up in the watershed groundwaters: i) reduces peak 1960's tritium concentrations by about 25%, ii) results in a 10% increase in 1970's tritium concentrations due to the delayed inflow of the weapons fallout peak; and iii) results in approximately 10% decrease in present and future predicted tritium concentrations. Increasing the mean residence time from 9 years to 24 years resulted in no more than a few percent change in model predictions. The scenario for mean residence time of 9 years provided the best fit to the measured data and was used in further predictions.

Cosmogenic & Weapons Fallout Source Term: Tritium concentrations at monitoring stations in the southern half of the Great Lakes Basin, Chicago (41.8N) and Madison (43.1N) were approximately 20% less on average than at the Ottawa station (45.3N). This is consistent with the continental effect of decreasing tritium concentration with decreasing latitude in North America, (Stewart and Farnsworth, 1968). To account for the lower tritium concentrations in the southern half of the basin, concentrations of tritium in precipitation were reduced by factor 0.85 for the lake basins lying south of an east west line centered on Ottawa (Michigan, Huron, Erie and Ontario).

The reduction in tritium input reduces lake concentrations by factors of 0.85 to 0.88. Predictions for Lake Huron with the modified input function with delay in watersheds are shown in Figure 3. Considering all lakes, the ratios of predicted to measured peak tritium concentrations range from 1.1 to 1.16. The ratios of predicted to measured tritium concentrations for all the data ranged from 0.62 to 1.52. Overall, the modified weapons fallout source term provides improved predictions and was used for the predictions.

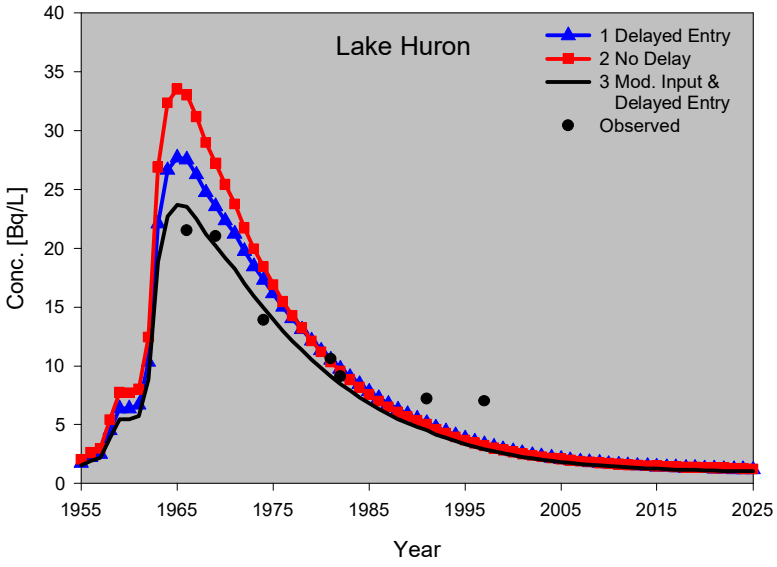

Figure 3 Predicted and observed tritium concentrations for Lake Huron. Predictions for cosmogenic and weapons fallout tritium only. Curve 1: Delayed entry of tritium from watershed; Curve 2: No delay in entry from lake watersheds; Curve 3: prediction for factor 0.85 reduction in cosmogenic-weapons fallout tritium for Lakes Michigan & Huron and delayed entry from watersheds.

Relative Contribution of the Cosmogenic, Weapons Fallout and CANDU Source Terms

The relative contribution of the three source terms to lake tritium levels is determined from use of the following source term combinations:

i) the cosmogenic source term alone,
ii) the cosmogenic and weapons source term, and
iii) all three source terms (cosmogenic, weapons fallout and CANDU) together.

Predicted lake tritium concentrations and the relative contribution of each source term to Lakes Huron and Ontario are shown for the period 1970 to 2025 (Figure 4). The figure shows the increasing contribution of CANDU tritium to the lakes since the start up of the reactors in 1970. Model projections for 1999 and beyond are based on 1996 CANDU emission data. These are considered most representative of future levels. The most recent emission data for 1997 and 1998 are considered to be low due to the temporary shutdown of 4 reactor units at both the Bruce and Pickering nuclear generating stations.

Model predictions are sensitive to the percentage CANDU airborne tritium emissions assumed to enter the lakes. Assuming 100% of the emissions enter the lakes, predictions rise by approximately 25%.

Considering all five Great Lakes for the period 1958 to 1997, good overall agreement between predicted and measured concentrations is observed. Predicted to measured ratios ranged from 0.62 to 1.87 and 67% of the model predictions were within 25% of the measured data. Measured and predicted values for the most recent (1997) lake tritium concentrations are listed in Table 3. For the 1997 data, the ratio of model calculated to measured values ranged from 1.06 to 1.35.

Computed cosmogenic, weapons fallout and CANDU tritium contributions to lake tritium inventories for 1999 are 6%, 30% and 64% for Lake Huron and 8%, 27% and 65% for Lake Ontario. Projections show that the relative contributions of CANDU tritium will increase to approximately 80% by 2025 while the contribution of weapons fallout tritium will decline to approximately 10%. This is due to the continued decay and outflow of weapons fallout tritium from the lakes. Lake Huron and Ontario concentrations are projected to decline from 1997 levels of 8.1 and 7.9 Bq/l to 5 and 4.9 Bq/L by 2025. It should be noted that despite the large contribution of CANDU tritium to the lakes, the lake concentrations are three orders of magnitude lower than the drinking water standard (International Joint Commission, 1977).

If all reactors were shut down in 1999, projected tritium concentrations in 2025 for Lakes Huron and Ontario are 1.2 and 1.3 Bq/L. Projected tritium concentrations in 2025 for no CANDU inputs to the lake ever (prior and post 1999) are 1.0 and 1.2 Bq/L for Lakes Huron and Ontario. The similarity between these two projections is due to the rapid removal of lake tritium inventories by water outflow from the lakes and radioactive decay.

Table 3 Measured and Model Calculated 1997 Great Lakes Tritium Concentrations

Lake	1997 Measured [Bq/L]	1997 Model Calculated [Bq/L]
Superior	2.0	2.7
Michigan	3.0*	3.1
Huron	7.0	8.1
Erie	5.5	5.8
Ontario	7.1	7.9

* 1998 data

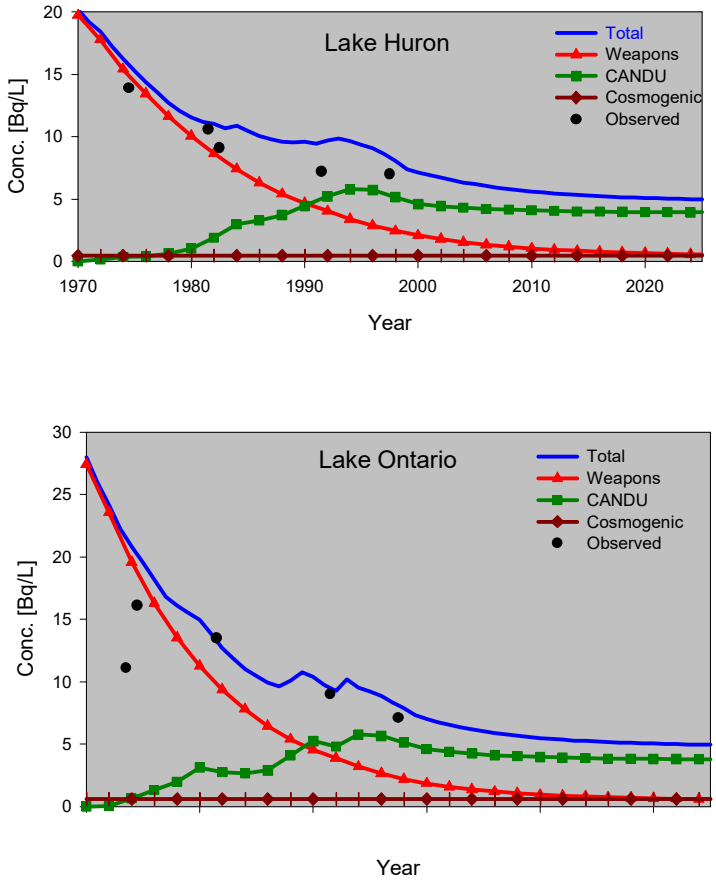

Figure 4 Predicted and observed tritium concentrations for Lakes Huron and Ontario projected to 2025. Predictions for total tritium and contributions of CANDU, weapons fallout and cosmogenic sources.

CONCLUSIONS

A concentration-time model has been used to evaluate the impact of CANDU tritium emissions on the Great Lakes. Uncertainties in the model are the residence time in lake watersheds, the weapons fallout source term and the amount of airborne CANDU emissions entering the lakes. The model was calibrated to determine the most appropriate values of the watershed residence time and weapons fallout source term. The conservative estimate was made that 50% of the airborne CANDU tritium emissions enter the lakes.

The relative contributions of CANDU tritium to present day Lakes Huron and Ontario inventories are 64 and 65% respectively. Assuming current CANDU tritium emissions continue into the future, the lake concentrations are projected to decline from current

levels of 7 Bq/L to 5 Bq/L by 2025 as residual weapons fallout tritium continues to be removed from the lakes by outflow and decay. The relative contribution of CANDU tritium to the total lake tritium inventories in Lakes Huron and Ontario are projected to increase to approximately 80% by 2025 as the inventory of the residual weapons testing tritium declines.

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