# A Model for Calculating Radionuclide Concentrations in the Fermilab Industrial Cooling Water System

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# Abstract

Large particle accelerators such as those at the Fermi National Accelerator Laboratory (Fermilab) unavoidably produce radionuclides in their bulk shielding and also require large volume process cooling water systems to handle electrical heat loads. The Neutrinos at the Main Injector (NuMI) beam facility came on line during CY 2005. After several months of initial operation of this facility, measurable concentrations of <sup>3</sup>H as tritiated water were identified throughout the Industrial Cooling Water (ICW) system. This was the first identification of measurable concentrations of any radionuclide leaving the Fermilab site, motivating an extensive effort to understand these discharges in order to minimize environmental releases. As part of this work, a simple model of radionuclide concentrations in this system has been developed.

Key Words: accelerator, surface water, environmental modeling, environmental transport

# Brief Description of the Fermilab Industrial Cooling Water System

The Fermilab ICW System is a network of ponds, connecting ditches, and underground pipelines along with ancillary equipment such as pumps, filters, aeration devices, and cooling towers. The system has been expanded several times during the 40-y lifetime of Fermilab to match the operational needs of an evolving, often growing, facility. Capacity enhancements have been especially important in enabling successful operations during the hot, humid summers of northern Illinois. Figs. 1 and 2 are aerial photographs and Fig. 3 is a line drawing of the Fermilab site showing the main features of this system.

The system provides non-contact secondary cooling of accelerator components, withdrawing heat from a much smaller low conductivity water (LCW) system that circulates directly through accelerator components. The smaller LCW system is designed to meet stringent purity requirements necessary for proper functioning of equipment. Prior to 2005 input to the ICW system consisted of natural precipitation, pumping from on-site wells, and state-permitted withdrawals from the Fox River. This river is about 4.8 km west of the Fermilab site. Withdrawals are permitted only when the flow rate of the river exceeds a specified value. Output from the system consists of evaporation, both natural and in cooling towers, and permitted discharges to three small creeks.

The Neutrinos at the Main Injector (NuMI) facility has been discussed elsewhere (Hylen et al. 1997; Cossairt 1998; Abramov et al. 2002). It includes a target, a large decay pipe, and a high beam power hadron absorber all located in an underground cavern. At present 200-400 kW of 120 GeV protons are delivered to it. By design, this beamline produces radionuclides in its environs that unavoidably become co-mingled with the outflow of tunnel dewatering and can be said to function essentially as a large well. The outflow is  $1.104 \times 10^{-2} \text{ m}^3 \text{s}^{-1}$  (about 175 gallons min<sup>-1</sup>) of relatively cool (about 12 °C), clear water. It is highly suitable for secondary cooling and was added to the ICW system.

Table 1 gives some relevant facts about the cooling pond system and water balance parameters. The values vary significantly both seasonally and from year-to-year due to meteorological conditions. The approximate volumes for both FULL ponds and LOW ponds (drought conditions) are provided. Also given are average volume flow rates of water entering the system, the maximum intake from the Fox River, and the intake from the wells including NuMI. If the pond volume *V* is constant, the input flows are balanced by the water leaving the pond system by means of evaporation, discharges to the three creeks, and infiltration into the ground. Otherwise the input either exceeds the output when the ponds are filling or is less than the output when the ponds are shrinking. Values are provided for "summer" (higher) and "winter" (lower) precipitation rates. The values are representative ones, spanning the domain of the possibilities between those involved when the ponds are full, the average summer precipitation rate is present, and the maximum Fox River intake is available; and winter conditions where the ponds are low, no precipitation is present, and the water from Fox River is not available ("drought" conditions). The latter is perhaps an extremum; due to the distance from its headwaters in southeastern Wisconsin, the Fox River withdrawal is sometimes available during local drought conditions.

#### **Model Assumptions**

- The ICW system volume V is a constant in time; dV/dt = 0. This is reasonable for time periods on the order of weeks. Including expanding or contracting ponds in the model considered here would require the solution of a *partial* differential equation; not deemed necessary for present purposes.
- As evidenced by recent experience, the water in the pond system can be "manipulated" (i.e., mixed) so that the concentration of radionuclides in the water is a constant throughout the system. The model presented here can also be adapted to a subset of the ICW system.
- Surface conditions do not affect the delivery of radionuclides from small sump discharges from accelerator enclosures. This likely overestimates the delivery of radionuclides during drought conditions because many of these sumps go dry in such circumstances.

#### **Turnover Rates of Water**

The water turnover mean-life of the pond system  $\tau_{ponds}$  is the system volume divided by the total flow rate of the chosen scenario. The corresponding clearance rate  $\lambda_{ponds}$  is;

$$\lambda_{ponds} = \frac{1}{\tau_{ponds}} \,. \tag{1}$$

Doing this calculation for selected scenarios leads to the values in Table 2. The unit of time is chosen as days (d) for practical convenience. The "intermediate" scenarios represent an average of summer and winter precipitation values likely encountered over long periods of accelerator and ICW system operations.

#### Calculating the Concentration in the Pond System

#### **Calculated Concentration in Water Released to the Surface**

The so-called Concentration Model (CM) is the current methodology used at Fermilab to estimate concentrations delivered to surface water and groundwater (Malensek 1993). In this model it is assumed that an irradiation due to beamline operations at constant beam intensity has been carried out for a time t after an initial startup with no radionuclides initially present. After such an irradiation the concentration

 $s_j(t)$  of the  $j^{th}$  radionuclide delivered to the surface will be given by the following solution to the activation equation including mixing (e.g., Cossairt 2007):

$$s_{j}(t) = \frac{\lambda_{j}}{\lambda_{j} + r} P_{j} \left\{ 1 - \exp\left[ -\left(\lambda_{j} + r\right) t \right] \right\},$$
<sup>(2)</sup>

where  $\lambda_j$  is the physical decay constant of the *j*<sup>th</sup> radionuclide (the reciprocal of the physical mean-life  $\tau_j$ ) and the factor  $P_j$  includes the parameters related to the calculation of the *static* concentration that would occur with no water movement during the irradiation. The parameter *r* is the rate at which the water turns over *within* the volume of activation. Eq. (2) agrees with intuition; for a negligible turnover rate ( $r << \lambda_j$ ), one is left with the *static* concentration while for rapid turnover,  $r >> \lambda_j$ , the concentration is greatly reduced. Of course, under conditions of no flow, there would no discharge. An added potential element of complexity not directly addressed here is that of the "source" consisting of a superposition of multiple sources with individual values of *r*. Eq. (2) applies to media where the mixing readily occurs. For regions where the flow is sufficiently slow through the media to allow activation to occur at different rates in various locations within the activation zone, the solution to a related partial differential equation is needed. An elementary example is given by Cossairt (2007).

#### **Dilution in the ICW System Concurrent with Operations**

One begins by defining  $\lambda_{source}$ , the reciprocal of the mean-life for a given source to fill the ICW system as the sole input if  $f_{source}$  is the output flow rate of the source in question;

$$\lambda_{source} = \frac{f_{source}}{V} \,. \tag{3}$$

Continuing to assume that dV/dt = 0;

$$\frac{dC_{j}(t)}{dt} = \lambda_{source} s_{j}(t) - \lambda_{ponds} C_{j}(t) - \lambda_{j} C_{j}(t) , \qquad (4)$$

where  $C_j$  is the concentration in the ICW of the  $j^{th}$  radionuclide as a function of run time *t*. The left-hand side is equal to the sum of three terms. The first represents the delivery of the  $j^{th}$  radionuclide in the water

being discharged from the source  $s_j$  into the pond system. The second term is the flow of water having concentration  $C_j$  out of the system. The third is the physical decay of the radionuclide of interest.

Eq. (4) is more closely aligned with a standard form if rewritten as follows:

$$\frac{dC_{j}(t)}{dt} + \left(\lambda_{j} + \lambda_{ponds}\right)C_{j}(t) = \lambda_{source}s_{j}(t).$$
(5)

Combining constants and rewriting this equation will be helpful;

$$\frac{dy}{dx} + ay = b(1 - e^{-dx}), \tag{6}$$

where  $a = \lambda_j + \lambda_{ponds}$ ,  $b = \frac{\lambda_{source} \lambda_j P_j}{\lambda_j + r}$ , and  $d = \lambda_j + r$ .  $C_j$  has been replaced with y and t with x. Then

$$\frac{dy}{dx} + P(x)y = Q(x).$$
<sup>(7)</sup>

Using an integrating factor F and letting K be an arbitrary constant determined from the initial condition of a concentration of zero (y=0) at zero time (x=0), then solving for y:

$$F = \int P(x)\delta x$$
, here  $F = \int a\delta x = ax$ , then (8)

$$ye^{F} = \int e^{F}Q(x)\delta x + K \to ye^{ax} = b \int e^{ax} (1 - e^{-dx})\delta x + K = \frac{be^{ax}}{a} - \frac{be^{(a-d)x}}{a-d} + K.$$
(9)

$$y = \frac{b}{a} - \frac{be^{-dx}}{a-d} + Ke^{-ax},$$
 (10)

$$K = \frac{b}{a-d} - \frac{b}{a}, \text{ and}$$
(11)

$$y = \frac{b}{a} \left( 1 - e^{-ax} \right) + \frac{b}{a - d} \left( e^{-ax} - e^{-dx} \right).$$
(12)

Checking the results:

$$\frac{dy}{dx} = be^{-ax} - \frac{ab}{a-d}e^{-ax} + \frac{bd}{a-d}e^{-dx}, \text{ and } ay = b - be^{-ax} + \frac{ab}{a-d}e^{-ax} - \frac{ab}{a-d}e^{-dx}, \text{ so that}$$

$$\frac{dy}{dx} + ay = b + \left(\frac{bd}{a-d} - \frac{ba}{a-d}\right)e^{-dx} = b\left(1 - e^{-dx}\right).$$

For *x*=0, *y*=0 so the initial condition is verified. Reinserting the constants and variables;

$$C_{j}(t) = \frac{\lambda_{source} \lambda_{j} P_{j}}{(\lambda_{j} + r) (\lambda_{j} + \lambda_{ponds})} \Big[ 1 - \exp\{-(\lambda_{j} + \lambda_{ponds})t\} \Big]$$

$$+ \frac{\lambda_{source} \lambda_{j} P}{(\lambda_{j} + r) (\lambda_{ponds} - r)} \Big[ \exp\{-(\lambda_{j} + \lambda_{ponds})t\} - \exp\{-(\lambda_{j} + r)t\} \Big]$$

$$(13)$$

The second term, a "transient", becomes negligible after a period of time, but it does have a nonphysical singularity in the unlikely coincidence that *r* should be equal to  $\lambda_{ponds}$ . If the equilibrium value of  $s_j$  is known, e.g., from measurement, one can use it in Eq. (13) in place of the factor  $\lambda_j P_j / (\lambda_j + r)$ .

#### **Clearance from the Pond System Subsequent to Operations**

Assuming for simplicity that the beam is operated at a constant intensity during a "run" of some time duration  $t_{run}$ , then the concentration being delivered to the ICW system by a single delivery mechanism at the end of this period will be

$$s_{j}(t_{run}) = \frac{\lambda_{j}}{\lambda_{j} + r} P_{j} \left\{ 1 - \exp\left[ -\left(\lambda_{j} + r\right) t_{run} \right] \right\}.$$
(14)

Likewise, at the end of this operational period, from Eq. (13) the concentration in the pond system will have a value  $C_j(t_{run})$  for  $t=t_{run}$ . At the end of operations, radioactivity is no longer being produced but will continue to be delivered by the source to the ICW system in accordance with the values of  $\lambda_j$  and r. Here "primed" variables (e.g., t') will be used to distinguish those that might have different values during the non-operational period such as the water turnover rate. Taking t' as the time since the cessation of operations, the concentration of radionuclide j delivered into the ICW system will now have the following time dependence, making the assumption that the absence of operations makes no difference in the rate of removing water from the activation zone:

$$s'_{j}(t') = s_{j}(t_{run}) \exp\left[-(\lambda_{j}+r)t'\right].$$
(15)

It is clear that the relevant differential equation is

$$\frac{dC_{j}(t')}{dt} + \left(\lambda_{j} + \lambda'_{ponds}\right)C_{j}(t') = \lambda_{source}s'_{j}(t').$$
(16)

The algebra will again be easier if the constants are merged as before and a standard form is used;

$$a' = \lambda_j + \lambda'_{ponds}, \ b' = \lambda_{source} s_j(t_{run}), \text{ and } \ d = \lambda_j + r$$
 (17)

The same technique will be used to solve Eq. (16) as was employed to solve Eq. (5);

$$\frac{dy}{dx} + a'y = b'e^{-dx}.$$
(18)

The integration factor is  $F = \int a' \delta x = a' x$ . Applying it results in:

$$ye^{a'x} = b'\int e^{a'x}e^{-dx}\delta x = b'\int e^{(a'-d)x}\delta x + K' = \frac{b'e^{(a'-d)x}}{a'-d} + K', \text{ and}$$
 (19)

$$y = \frac{b'}{a - d'}e^{-dx} + K'e^{-ax}.$$
 (20)

This time the initial condition is  $y=C_j(t_{run})$  for x=0, so

$$K' = C_j(t_{run}) - \frac{b'}{a' - d} \quad \text{and} \tag{21}$$

$$y = C_{j}(t_{run})e^{-a'x} + \frac{b'}{a'-d} \left(e^{-dx} - e^{-a'x}\right).$$
 (22)

Checking the solution again and verifying the initial condition;

$$\frac{dy}{dx} = -a'C_{j}(t_{run})e^{-a'x} - \frac{b'd}{a'-d}e^{-dx} + \frac{a'b'}{a'-d}e^{-a'x} \text{ and } a'y = a'C_{j}(t_{run})e^{-a'x} + \frac{a'b'}{a'-d}e^{-dx} - \frac{a'b'}{a'-d}e^{-a'x}, \text{ so that}$$
$$\frac{dy}{dx} + a'y = \frac{a'b'}{a'-d}e^{-dx} - \frac{b'd}{a'-d}e^{-dx} = b'e^{-dx}.$$

Substituting in the parameters and variables;

$$C_{j}(t') = C_{j}(t_{run}) \exp\left[-\left(\lambda_{j} + \lambda'_{ponds}\right)t'\right] + \frac{s_{j}(t_{run})}{\lambda'_{ponds} - r} \left[\exp\left\{-\left(\lambda_{j} + r\right)t'\right\} - \exp\left\{-\left(\lambda_{j} + \lambda'_{ponds}\right)t'\right\}\right].$$
(23)

The first term represents the decline of the concentration as a function of the time since beam line operations ceased. The second term takes into account the ongoing effect of additional radioactivity coming in if that persists for some significant time period compared with the other time constants involved. A nonphysical singularity results in the unlikely coincidence that  $r = \lambda'_{nonds}$ .

#### **Successive Periods of Operation and Multiple Source Components**

The contributions of successive operational periods to the total activity found at any point in time are additive. Following a period of operations, the concentrations will be built up to some value. Then, during a subsequent shutdown period, the water turnover described will reduce the concentration present according to the pond and source parameters applicable at that time. Resuming operations will start delivering radionuclides in the pond system again, superimposing additional content on top of what remains from previous operational periods, governed by the seasonal and operational time constants. This could be evaluated by taking a "step-wise" approach. In real-life situations, sources may be comprised of multiple components with different associated time constants with results being additive.

#### An Example from the Initial Operation of the NuMI Facility

The initial operational period of the NuMI facility provides an opportunity to test this model. In these calculations, the two values of  $\lambda_{ponds}$  from Table 2 for "Intermediate" seasonal conditions when the ponds are FULL and LOW will be used, since high energy physics operational periods span multiple climatic seasons and it is known that the initial months of NuMI operation included both periods of normal rainfall and several months of significant, indeed near record, drought. The condition of the ponds being FULL makes the assumption that the Fox River intake is turned ON while the condition of the ponds being LOW takes the Fox River intake as being OFF. The corresponding values of  $\lambda_{source}$ (FULL)=1.02x10<sup>-3</sup> d<sup>-1</sup> and  $\lambda_{source}$ (LOW)=1.42x10<sup>-3</sup> d<sup>-1</sup> are obtained using the near-constant discharge from the NuMI dewatering of 1.104 x 10<sup>-2</sup> m<sup>3</sup>s<sup>-1</sup> (954 m<sup>3</sup>d<sup>-1</sup>). The calculations do not include any directly evaporated tritiated water that does not enter the pond system. For <sup>3</sup>H,  $\lambda_{j}$ =1.540x10<sup>-4</sup>d<sup>-1</sup>. Unfortunately, the parameter *r* has to be estimated. From data accumulated during 2006-2007, it is clear that there are

likely multiple sources delivering tritiated water within the NuMI facility (Pordes 2007; Finsterle et al. 2007). It appears that the effective mean-life of the most rapid-clearing component of the concentrations of tritiated water in the NuMI sump discharge is of the order of 3 days. This corresponds to a value of r of about 0.3 d<sup>-1</sup> and is called the *fast* component. There is also a *slow* component with a mean-life of several hundred days or longer, corresponding to an approximate value of r of 0.003 d<sup>-1</sup>.

Figs. 4-6 show results for specified conditions each with 3 choices of the value of *r*. The results for the conditions studied were all insensitive to values of *r*>0.3 d<sup>-1</sup>. Conditions of both FULL and LOW ponds are considered to bracket the possible results. Figs. 4 and 5 calculate the buildup of concentrations of tritium in the ponds during operational periods. In each graph the "Fractional Concentration" is the ratio  $C_j/s_j$ . Figs. 6 and 7 calculate the decline of the concentrations of tritium in the ponds during shutdown periods for the indicated pond system scenario and give the "Fractional Concentration" measured against the concentration in the ponds at the beginning of the shutdown period. For the beam ON scenarios, the characteristic time constant  $\tau$  required to reach 63.2 % (1-1/e) of that equilibrium value is given. For the beam OFF scenarios, the characteristic time constant  $\tau$  required to reach 36.8 % (1/e) of the initial value of the fractional concentration is provided.

A reasonable scenario for approximately the first 120 days of operation of NuMI at high intensity in late summer and autumn of 2005 is that of "Precipitation INTERMEDIATE, Beam ON, Ponds LOW, and Fox River Intake OFF". During this operational period the concentration in the NuMI discharge was typically  $9.25 \times 10^5$  Bq m<sup>-3</sup>. From the calculation that generated Fig. 5, one gets 3 *r*-dependent values of the fractional concentration of tritium in the ICW that are given in Table 3. Averaging the results of a site wide sampling of the ponds conducted on 30 November 2005 resulted in an average concentration of  $8.9 \times 10^4$  Bq m<sup>-3</sup>. This value is consistent with the above results if the more rapid source components are dominant. Subsequently, increased precipitation levels and improved water management practices rendered the tritium concentrations generally to levels below those detectable.

# **Conclusion and Acknowledgments**

9

This model provides a physical understanding of delivery of radionuclides into the cooling ponds that should prove to be useful for future estimates. The author appreciates helpful discussions with S. Krstulovich, R. Walton, V. Cupps, P. Kesich, K. Vaziri, R. Plunkett, and W. Griffing. This work was supported by the U. S. Department of Energy, Office of Science under contract with Fermi Research Alliance, LLC.

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# Tables

ICW Volume FULL:	9.39 x 10 <sup>5</sup> n	n <sup>3</sup>				
ICW Volume LOW:	6.78 x 10 <sup>5</sup> n	$n^3$				
Season, Fox River and well status			Flow rates $(m^3 s^{-1})$			
Season	River	Wells	Precipitation	River	Wells	Total
Summer	ON	ON	0.076	0.050	0.054	0.180
Summer	OFF	ON	0	0	0.054	0.054
Winter	ON	ON	0.024	0.050	0.054	0.128
Winter	OFF	ON	0	0	0.054	0.054

Table 1 ICW system volumes and water input rates<sup>a</sup>

<sup>a</sup> S. Krstulovich and R. Walton, private communication, 2006.

Scenario			Water Mean-Lives and Turnover Rates			
Season	Ponds	River and precipitation status	Water mean- life, $\tau_{ponds}$ (s)	Water mean- life $\tau_{ponds}$ (d)	Water turnover rate $\lambda_{ponds}$ (d <sup>-1</sup> )	
Summer	FULL	ON	$5.20 \times 10^{6}$	60.1	1.66 x 10 <sup>-2</sup>	
Summer	FULL	OFF	$1.75 \ge 10^7$	203.0	4.93 x 10 <sup>-3</sup>	
Summer	LOW	OFF	$1.27 \ge 10^7$	147.0	6.82 x 10 <sup>-3</sup>	
Winter	FULL	ON	$7.32 \times 10^6$	84.7	1.18 x 10 <sup>-2</sup>	
Winter	FULL	OFF	$1.75 \times 10^7$	203.0	4.93 x 10 <sup>-3</sup>	
Winter	LOW	OFF	$1.27 \times 10^7$	147.0	6.82 x 10 <sup>-3</sup>	
Intermediate	FULL	ON	6.24 x 10 <sup>6</sup>	72.2	1.38 x 10 <sup>-2</sup>	
Intermediate	LOW	OFF	$1.27 \times 10^7$	147.0	6.82 x 10 <sup>-3</sup>	

Table 2 ICW System mean-lives and turnover rates

$r (d^{-1})$	Fractional concentration	Estimated concentration		
		$({\rm Bq} {\rm m}^{-3})$		
0.3	0.113	$1.04 \times 10^5$		
0.03	0.091	$8.42 \times 10^4$		
0.003	0.021	$1.92 \times 10^4$		

Table 3 Estimated ICW <sup>3</sup>H concentrations following 120 days of NuMI operations

# **List of Figure Captions**

- Aerial photograph from the southwest of the Fermilab site showing major components of the Fermilab ICW system. The photograph was provided by Fermilab Visual Media Services.
- Overhead photograph of the Fermilab site showing above-grade components of the Fermilab ICW system with system features shown in enhanced blue color. The photograph was provided by Fermilab Facilities Engineering Services.
- 3. Schematic diagram of the above grade components of the ICW system. The diagram was adapted from one provided by P. Kesich.
- 4. Calculated fractional concentrations of <sup>3</sup>H in the ICW system during NuMI operations with full ponds, intermediate precipitation levels, and withdrawals from the Fox River present. The approximate time constants inferred from the results are also given.
- 5. Calculated fractional concentrations of <sup>3</sup>H in the ICW system during NuMI operations with low ponds, intermediate precipitation levels, and withdrawals from the Fox River not present. The approximate time constants inferred from the results are also given.
- 6. Calculated fractional concentrations of <sup>3</sup>H in the ICW system subsequent to NuMI operations with full ponds, intermediate precipitation levels, and withdrawals from the Fox River present. The approximate time constants inferred from the results are also given.
- Calculated fractional concentrations of <sup>3</sup>H in the ICW system subsequent to NuMI operations with low ponds, intermediate precipitation levels, and withdrawals from the Fox River not present. The approximate time constants inferred from the results are also given.

Fig. 1



Fig. 2



Fig. 3





Fig. 5





Fig. 7

