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A Model for Calculating Radionuclide Concentrations in Fermilab Cooling Ponds

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1. Introduction

The goal of this paper is to describe a methodology for calculating the concentrations of radionuclides in the Fermilab cooling pond system resulting from discharges to these impoundments. This work is motivated by two purposes. The first is to achieve a better understanding of the effect of the surface water discharge from the Neutrinos at the Main Injector (NuMI) beamline during its initial run that began in CY 2005 and concluded in late February 2006. The second is to provide input to a possible policy revision that might serve to improve the control of radionuclide discharges into surface waters at Fermilab. It is unlikely that radionuclides aside from tritium (^3H) are of importance. However, the discussion in this paper has been generalized to include multiple radionuclides. This work is complementary to and supportive of that performed by S. Krstulovich who has devised a detailed Microsoft Excel™ spreadsheet model of the cooling ponds and industrial cooling water (ICW) system at Fermilab.

2. Facts about the cooling pond system at Fermilab

Table 1 gives some relevant facts about the cooling pond system and water balance parameters. The approximate volumes for both "full" ponds and "low" ponds (drought conditions) are provided. Also given are average time rates at which water enters the system including seasonal precipitation rates under "normal" meteorological conditions; the maximum intake from the Fox River; and the intake from the wells, inclusive of the present delivery of approximately 175 gallons min^{-1} from the NuMI sump discharge.[1] If the pond volume, V, is kept constant, then these 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 these input flows either exceed the output flows when the ponds are filling or are less than the output flows when the ponds are shrinking. Two sets of values are provided for "summer" and "winter" precipitation rates. The intent of this table is to list a set of representative values spanning the domain of the possibilities between those involved when the ponds are full, the average summer (larger) 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 Fox River is not available ("drought" conditions). The values in this table will be used to calculate water exchange rates under various scenarios.

Assumptions

In the course of the analysis presented here, several assumptions will be employed. These are:

[1] The values in Table 1 were obtained from discussions with S. Krstulovich and R. Walton and their help is much appreciated.

A. The pond system volume, V, is a constant in time, that is; $dV/dt = 0$. This is probably reasonable at least for time periods on the order of weeks given the large volumes present. That is, the rate of water being added is balanced by the rate of water being lost from the system. Including expanding or contracting ponds in the model considered here would require the solution of a more complicated *partial* differential equation; a step thought not to be necessary for present purposes.

B. 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. Present experience makes this simplification credible, but rather challenging to achieve. The model discussed here could be modified for application to a subset of the system where this condition might be more readily accomplished.

C. It is assumed that surface conditions do not affect the delivery of radionuclides from sump discharges. This choice overestimates the delivery of radionuclides during drought conditions because the sumps of enclosures located in the glacial till also sometimes go dry during droughts.

Table 1 Pond Volumes and Water Input Rates

Pond Volume FULL:	2.48×10^8	gallons				
Pond Volume LOW:	1.79×10^8	gallons				
Season, Fox River, and Well Status			**Flow Rates (gpm)**			
Season	**River**	**Wells**	**Precipitation**	**River**	**Wells**	**Total**
Summer	ON	ON	1210	800.0	850	2860
Summer	OFF	ON			850	850
Winter	ON	ON	380	800.0	850	2030
Winter	OFF	ON			850	850

Turnover Rates of Pond Water

One can calculate a water turnover mean-life of the pond system, here denoted τ_{ponds}, simply by dividing the system volume by the total flow rate of a chosen scenario. It follows that the clearance rate, here denoted λ_{ponds}, is given by

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

Doing this calculation for some scenarios leads to the values in Table 2. The two "intermediate" scenarios represent an average of summer and winter precipitation values that might be encountered over a long period of operations. In this table, the values for LOW ponds include only the input from the wells and the NuMI sump discharge given the desirability to retain water by all available means during drought, or near-drought, conditions. In doing so, the tacit, and not always correct, assumption is made that the Fox River water is unavailable during periods of drought of Fermilab.[2]

Table 2 Pond Water System Mean-Lives and Turnover Rates

Scenario			Water Mean-Lives and Turnover Rates		
Season	**Ponds**	**River and Precipitation Status**	**Water Mean-Life, τ_{ponds} (minutes)**	**Water Mean-Life τ_{ponds} (days)**	**Water Turnover Rate λ_{ponds} (per day)**
Summer	FULL	ON	8.66×10^4	60.1	1.66×10^{-2}
Summer	FULL	OFF	2.92×10^5	203.0	4.93×10^{-3}
Summer	LOW	OFF	2.11×10^5	147.0	6.82×10^{-3}
Winter	FULL	ON	1.22×10^5	84.7	1.18×10^{-2}
Winter	FULL	OFF	2.92×10^5	203.0	4.93×10^{-3}
Winter	LOW	OFF	2.11×10^5	147.0	6.82×10^{-3}
Intermediate	FULL	ON	1.04×10^5	72.2	1.38×10^{-2}
Intermediate	LOW	OFF	2.11×10^5	147.0	6.82×10^{-3}

3. Calculating the Concentration in the Pond System

Calculated Concentration Released to Surface Water

The Concentration Model (CM) is the current methodology used at Fermilab to estimate concentrations delivered to surface water and groundwater (Co99). Delivery to the groundwater is not within the scope of the present discussion. The CM allows one to average over a 3 year period of operations. This triennial averaging will not be used here as it is likely to be inappropriate for surface water discharges because these discharges can be sensitive to short-term events such as major rain storms. Assume 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. If the water is released to the surface at all, it may be reasonable to assume that the concentration within the spatial volume bounded by the attenuation of the radiation field to 1 % of its maximum value (the so-called "99% volume" of the CM) can be taken to be uniform due to rather rapid mixing. After such an irradiation time at an assumed constant beam intensity, the concentration, $s_j(t)$, of the j^{th} radionuclide delivered to the surface will be given by the following solution to the activation differential equation including mixing given, for example, in (Co06):

$$s_j(t) = \frac{\lambda_j}{\lambda_j + r} P_j \left\{1 - \exp\left[-\left(\lambda_j + r\right)t\right]\right\}, \tag{2}$$

[2] The headwaters of the Fox River are in southeastern Wisconsin so this body of water can have a large flow at the Fermilab water intake concurrent with local droughts at the Fermilab site.

where λ_j is the physical decay constant of the j^{th} radionuclide (the reciprocal of the physical mean-life[3], τ_j) and the factor P_j includes the parameters related to the calculation of the *static* concentration in the concentration model assuming that no movement occurs during the irradiation. The parameter r is the rate at which the water turns over *within* the 99% volume[4]. This formula agrees with intuition if one realizes that for a negligible turnover rate ($r << \lambda$), one is left with the *static* concentration (the concentration that would be present if the water did not move) while for rapid turnover, $r >> 1$, the concentration is greatly reduced. Of course, under conditions of no flow, there should be no delivery of radionuclides to the ponds, either. An added element of complexity not completely addressed by this paper is the fact that a given "source" may consist of the superposition of multiple "sources", or delivery mechanisms, each with its own value of r, with a resulting additional time-dependence.

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 more complicated, but related, partial differential equation is needed. An example is given in (Co06).

Dilution in the Pond System

Continuing to make the simplifying assumption that the system volume, V, is constant with time; in other words, $dV/dt = 0$; one has

$$\frac{dC_j(t)}{dt} = \frac{f_{source}}{V} s_j - \frac{f_{ponds}}{V} C_j(t) . \tag{3}$$

In Eq. (3), C_j is the concentration in the pond system of the j^{th} radionuclide as a function of run time, t. Its derivative with respect to time (the left-hand side) is equal to the sum of two terms given on the right-hand side. The first term is the rate of volume flow of the source of radionuclides, f_{source}, times the concentration of the j^{th} radionuclide in the water being discharged from the source, s_j, *into* the pond system. The second term represents the flow of water having concentration C_j *out of* the system at a total flow rate, f_{ponds}. In this equation it is assumed that the physical lifetime of the radionuclide is much longer than the time constants of water exchange within the pond system and that the value of s_j rises to the equilibrium value implied by Eq. (2) with a time constant that is short compared with those associated with the turnover of water in the pond system or with the decay of the radionuclide and is taken to be a constant. This assumption is often sensible for long-lived radionuclides exemplified by ^3H with its 12.32 year half-life (τ_j = 17.77 yr) for typical "sources" at Fermilab. A more complete mathematical solution derived without these assumptions is presented in Appendix A. It is clear that

$$\lambda_{ponds} = \frac{f_{ponds}}{V} . \tag{4}$$

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[3] The physical mean-life, τ_j, is related to the more commonly tabulated physical half-life, $t_{j,1/2}$ via $\tau_i = (t_{j,1/2})/\ln 2$.

[4] The choice of the so-called "99 %" volume is admittedly arbitrary. Alternative choices of the "activation zone" are, of course, feasible.

Similarly, the coefficient of s_j in Eq. (3) is just the reciprocal of the mean time required for the individual source to fill the pond system all by itself and represents the time rate of accomplishing this, denoted λ_{source}, and given by

$$\lambda_{source} = \frac{f_{source}}{V}. \quad (5)$$

Thus, Eq. (3) can be written as follows:

$$\frac{dC_j(t)}{dt} = \lambda_{source} s_j - \lambda_{ponds} C_j(t). \quad (6)$$

After a time period of operations, t, if we have the initial condition that $C_j = 0$, the solution to Eq. (6) is

$$C_j(t) = \frac{\lambda_{source} s_j}{\lambda_{ponds}} \left[1 - \exp\left(-\lambda_{ponds} t\right)\right]. \quad (7)$$

Examples related to NuMI operations using the equation and its more complicated analog derived in Appendix A are provided in Appendix B.

4. Predictions of the Clearing of Radionuclides from Pond System Following Cessation of Operations

The clearing of radionuclides from the pond system will be determined by the time constants associated with both the pond system and the mechanisms that result in the release of radionuclides to them. For simplicity, one can sometimes assume that the contribution of the source ceases immediately. After a run of time duration t_{run}, the concentration will have built up to a value $C_j(t_{run})$, as determined above. This could be for a single period of operations or following the accumulation of multiple operational periods. During such a non-operational period, the clearance rate of water from the system is characterized by the parameter λ'_{ponds} that *could* be different from the λ_{ponds} that might characterize this phenomenon during the preceding operational period. Without further significant contributions from any sources, the concentration in the pond system will be given by

$$C_j(t') = C_j(t_{run}) \exp\left[-\left(\lambda_j + \lambda'_{ponds}\right)t'\right], \quad (8)$$

where the physical decay constant, λ_j, has been explicitly included in recognition of the fact that during non-operational periods during droughts, λ_j *could* be more comparable in magnitude to λ'_{ponds}. In Eq. (8), t' is used to measure the time during the non-operational period since operations ceased. In other words, $t' = 0$ at the beginning of the shutdown. Appendix A contains a more detailed description obtained without making some of these simplifying assumptions. Illustrative results related to the recent experience with NuMI are presented in Appendix B.

5. Successive Periods of Operation and Multiple Source Components

It should be noted that the contributions of successive operational periods to the total activity found at any point in time are additive. In other words, following a period of operations, the concentrations will be built up to some value as discussed above. Then, during a subsequent shutdown period, the water turnover described in Section 4 will serve to reduce the concentration present according to the pond and source parameters applicable at that time. However, this will likely not be to "zero" concentration. Resuming operations will start delivering radionuclides in the pond system again, superimposing additional content on top of what remains from previous operational periods. This cycle will be governed by the seasonal and operational time constants and could, in principle, be evaluated in detail by taking a "step-wise" approach. It should also be noted that sources may be comprised of multiple components with different associated time constants that will be additive.

6. Individual Sources in the "Cut and Cover" Zone

The intrinsic nature of accelerator operations results in a number of "loss points" in the accelerator lattice and also the external beamlines, as well as locations of deliberate beam targeting. Most of these occur in the "cut and cover" zone near the surface adjacent to accelerator/beamline components. Underdrains are used to drain the enclosures and the water is pumped to the surface as "sump discharges". One must be able to assess the collective impact of all of these sources on the total concentration in the cooling pond system. There are perhaps as many as 50 such locations within the Fermilab accelerator complex. It is believed that typical sump discharges in the Fermilab accelerator complex range from about 100 gallons day^{-1} to perhaps a few thousand gallons day^{-1}. Thus, the values of f_{source} for any of these sources, and perhaps even their sum, may be far less than the approximate value of 252,000 gallons day^{-1} represented by the NuMI sump discharge. The volumetric sum of these discharges may merit further study. However, under the present (May 2006) implementation of the CM (i.e., in FRCM Chapter 11), concentrations up to the DOE Derived Concentration Guides (DCGs) in designs (DOE90) are permitted by Fermilab policy in such discharges.[5]

Motivated by the experience with NuMI, it has become clear that measurable radionuclide concentrations in the ponds may occur if concentrations near the DCG were to be continuously dumped into a sump having a flow rate of "intermediate" size. Thus, for new designs, an estimate of the concentrations being released, s_j, should be calculated, a value of f_{source}, should be estimated, and the individual contribution to C_j should be calculated as described in this paper or by some alternative methodology. Given the known existence of a significant number of other, pre-existing, sources, it would be prudent to make sure that the design renders the value of C_j due to any particular source to be a very small fraction (e.g., perhaps < 1%) of some criteria, for example a specified, standardized limit of detection. Demonstration of such a value could be part of the normal shielding assessment review procedures embodied in *Fermilab Radiological Control Manual* (FRCM) Chapter 8. A plan to exceed this criterion in a design might warrant the requirement of a specific Director's exception following procedures set forth in the

[5] The presence of multiple radionuclides implies that the sum of the ratios of the concentrations of the individual radionuclides to their individual DCGs must be less than unity.

Fermilab Environment, Safety, and Health Manual (FESHM), a document which includes the FRCM.

A numerical example may illustrate how this might work. Assume a beam loss point generates water calculated to have an average value of $s_{tritium}$ = 100 pCi cm^{-3} from Eq. (2). The best available information may indicate that a reasonable estimate for the discharge rate, f_{source}, is 500 gallons day^{-1}. From the methodology presented here, one obtains value of λ_{source} = 2.8 x 10^{-6} day^{-1}; if the pond volume is conservatively assumed to be LOW. If one further assumes that this source will operate indefinitely under drought conditions and apply Eq. (7) with an "infinite" irradiation time, one gets

$$C_{tritium} = \frac{2.8\times10^{-6}\ \text{day}^{-1}}{6.84\times10^{-3}\ \text{day}^{-1}} 100\ \text{pCi cm}^{-3} = 0.04\ \text{pCi cm}^{-3}.$$

This result illustrates that only 25 such sources would need to exist to obtain a value of $C_{tritium}$ > 1 pCi cm^{-3}, the current limit of detection. Further discussions are needed to refine the nature of this or a similar criterion that may be incorporated into Laboratory policy. Obviously, it might be necessary for Fermilab management to prioritize the "allowable" discharges based upon programmatic priorities in some manner rather than to simply treat all locations of beam loss equally.

7. Conclusion

The model discussed in this note provides a physical understanding of delivery of radionuclides into the cooling ponds under some simplifying assumptions. In particular, an understanding of the time constants involved is described in a way that is amenable to more detailed modeling. A policy revision to control the total activity as well as the concentration of radionuclides being discharged into the Fermilab pond system appears to be desirable. To do this may require a better understanding of the myriad of sump discharges at Fermilab.

Acknowledgments

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References

(Co99) J. D. Cossairt, A. J. Elwyn, P. Kesich, A. Malensek, N. Mokhov, and A. Wehmann, "The Concentration Model Revisited, Fermilab Environmental Protection Note EP 17, June 1999. A synopsis of the Concentration Model is described in Chapter 11 of the *Fermilab Radiological Control Manual*.

(Co06) J. D. Cossairt, Chapter 8 in "Radiation Physics for Personnel and Environmental Protection", Fermilab Report TM-1834, Revision 9, November 2005.

(DOE90) U. S. Department of Energy Order 5400.5, "Radiation Protection of the Public and the Environment" (1990).

APPENDIX A
SOLUTIONS OF THE DETAILED DIFFERENTIAL EQUATIONS

Buildup of Concentration During Operations

For convenience, Eq (2), which gives the dependence of the source concentration due to a single delivery mechanism of a given radionuclide, s_j, on irradiation time, t is repeated here:

$$s_j(t_i) = \frac{\lambda_j}{\lambda_j + r} P_j \left\{1 - \exp\left[-(\lambda_j + r)t\right]\right\}. \tag{A1}$$

The relevant differential equation taking into account the finite physical decay constants, λ_j, for the concentration of this radionuclide in the pond system, C_j, is

$$\frac{dC_j(t)}{dt} = \lambda_{source} s_j(t) - \lambda_j C_j(t) - \lambda_{ponds} C_j(t). \tag{A2}$$

In Eq. (A2) the time rate of change of concentration in the pond system is equated with the sum of three terms. The first is the continual *addition* of the radionuclide to the system, the second is the ongoing *loss* of the radionuclide through its radioactive decay, and the third is the *loss* of the radionuclide through the water balance of the system including outflow and evaporation. The equation is more closely aligned with a standard form found in mathematics textbooks if rewritten as follows:

$$\frac{dC_j(t)}{dt} + (\lambda_j + \lambda_{ponds}) C_j(t) = \lambda_{source} s_j(t), \tag{A3}$$

where what is happening to the pond system is balanced with the contribution from the source.

Combining constants and rewriting this equation will be helpful;

$$\frac{dy}{dx} + ay = b(1 - e^{-dx}), \tag{A4}$$

where $a = \lambda_j + \lambda_{ponds}$, $b = \dfrac{\lambda_{source}\lambda_j P_j}{\lambda_j + r}$, and $d = \lambda_j + r$.

This is recognizable as an equation of a standard form found if one replaces C_j with y and t with x:

$$\frac{dy}{dx} + P(x)y = Q(x). \tag{A5}$$

Such equations can be solved through the use of an integrating factor, F, obtained as follows from an indefinite integral:

$$F = \int P(x)\delta x \text{ , here } F = \int a\delta x = ax. \tag{A6}$$

Applying this factor in the standard manner;

$$ye^F = \int e^F Q(x)\delta x + K \text{ ,}$$

$$\text{here } ye^{ax} = b\int e^{ax}\left(1 - e^{-dx}\right)\delta x + K = \frac{be^{ax}}{a} - \frac{be^{(a-d)x}}{a-d} + K, \tag{A7}$$

where K is an arbitrary constant, here to be determined from the initial condition of a concentration of zero ($y = 0$) in the pond system. Solving the above for y;

$$y = \frac{b}{a} - \frac{be^{-dx}}{a-d} + Ke^{-ax}, \tag{A8}$$

If the initial condition that $y = 0$ for $x = 0$ is applied, then

$$K = \frac{b}{a-d} - \frac{b}{a}. \tag{A9}$$

Substituting,

$$y = \frac{b}{a}\left(1 - e^{-ax}\right) + \frac{b}{a-d}\left(e^{-ax} - e^{-dx}\right). \tag{A10}$$

The results of this exercise should be checked to see if this is a solution to Eq. (A4) [6];

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

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

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

Furthermore, for $x = 0$, $y = 0$ so the initial condition is checked to be correct.

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6 At least, it was considered prudent to do so for this author who needed to shake some "rust" off of his knowledge of differential equations!

Reinserting the constants and variables for the present problem into Eq. (A10);

$$C_j(t) = \frac{\lambda_{source}\lambda_j P_j}{(\lambda_j + r)(\lambda_j + \lambda_{ponds})}\left[1 - \exp\left\{-(\lambda_j + \lambda_{ponds})t\right\}\right]$$
$$+ \frac{\lambda_{source}\lambda_j P}{(\lambda_j + r)(\lambda_{ponds} - r)}\left[\exp\left\{-(\lambda_j + \lambda_{ponds})t\right\} - \exp\left\{-(\lambda_j + r)t\right\}\right] \quad \text{(A11)}$$

The first term of Eq. (A11) is equivalent to Eq. (7) if one considers λ_j to be negligible due to the long half-life of the radioactivity compared with the other time rate parameters. The second term usually becomes negligible after a period of time, but it does have a "nonphysical singularity" in the unlikely event that r should happen to be equal to λ_{ponds}. If the equilibrium value of s_j is known directly, likely from measurement, one can substitute its value directly into Eq. (A11) in place of the factor:

$$\frac{\lambda_j P_j}{\lambda_j + r}.$$

Decay of Concentration During a Shutdown Period

If the beam has operated at a constant intensity (assumed for mathematical simplicity) during a "run" of some time duration, t_{run}, then the concentration being delivered to the pond system by a single delivery mechanism at the end of this period will be given by

$$s_j(t_{run}) = \frac{\lambda_j}{\lambda_j + r} P_j \left\{1 - \exp\left[-(\lambda_j + r)t_{run}\right]\right\}. \quad \text{(A12)}$$

Likewise, at the end of this operational period, the concentration in the pond system will have a value $C_j(t_{run})$ obtained from Eq. (A11) for $t = t_{run}$, or directly by measurement. At the end of operations, radioactivity is no longer being produced. If the "clock" is reset so that $t' = 0$ now corresponds to the time since the cessation of operations, the concentration of radionuclide j delivered into the pond system will now have the following time dependence, making the tacit assumption that the lack of operations makes no difference in the rate of removing water from the activation zone:

$$s'_j(t') = s_j(t_{run})\exp\left[-(\lambda_j + r)t'\right]. \quad \text{(A13)}$$

Here we will use "primed" variables to distinguish these from those employed for the calculations done during the irradiation for those parameters that might have different values during the non-operational period (e.g., the water turnover rate).

It is clear that the relevant differential equation is, once again;

$$\frac{dC_j(t')}{dt} + \left(\lambda_j + \lambda'_{ponds}\right)C_j(t') = \lambda_{source}s'_j(t'). \tag{A15}$$

The algebra will again be easier if the constants are merged as before;

$$a' = \lambda_j + \lambda'_{ponds}\text{, } b' = \lambda_{source}s_j(t_{run})\text{, and } d = \lambda_j + r. \tag{A16}$$

The same technique employed above will be used to solve Eq. (A15), which will now be recast as before;

$$\frac{dy}{dx} + a'y = b'e^{-dx}. \tag{A17}$$

The integration factor is $F = \int a'\delta x = a'x$. Thus;

$$ye^{a'x} = b'\int e^{a'x}e^{-dx}\delta x = b'\int e^{(a'-d)x}\delta x + K' = \frac{b'e^{(a'-d)x}}{a'-d} + K'\text{, and so} \tag{A18}$$

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

Thus,

$$y = \frac{b'}{a-d'}e^{-dx} + K'e^{-ax}. \tag{A19}$$

But this time, the initial condition is not $y = 0$ for $x = 0$. Instead, it will be $y = C_j(t_{run})$ for $x = 0$. In view of that fact,

$$K' = C_j(t_{run}) - \frac{b'}{a'-d}\text{ and so} \tag{A20}$$

$$y = C_j(t_{run})e^{-a'x} + \frac{b'}{a'-d}\left(e^{-dx} - e^{-a'x}\right). \tag{A21}$$

A check of this solution is, again, in order;

$$\frac{dy}{dx} = -a'C_j\left(t_{run}\right)e^{-a'x} - \frac{b'd}{a'-d}e^{-dx} + \frac{a'b'}{a'-d}e^{-a'x}$$

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

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

Eq. (A21) is thus a solution to Eq. (A17). Furthermore, for $x = 0$, $y = C_j\,(t_{run})$, as it should.

Substituting in the parameters,

$$\begin{aligned} C_j(t') &= C_j(t_{run})\exp\left[-\left(\lambda_j + \lambda'_{ponds}\right)t'\right] \\ &+ \frac{s_j(t_{run})}{\lambda'_{ponds} - r}\left[\exp\left\{-\left(\lambda_j + r\right)t'\right\} - \exp\left\{-\left(\lambda_j + \lambda'_{ponds}\right)t'\right\}\right]. \end{aligned} \tag{A22}$$

The first term represents the decline of the concentration as a function of the time since beam line operations ceased and is that embodied in Eq. (8) of the main text. 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. It does become zero after a period of time. Again, a "nonphysical singularity" results if r should happen to be equal to λ'_{ponds}.

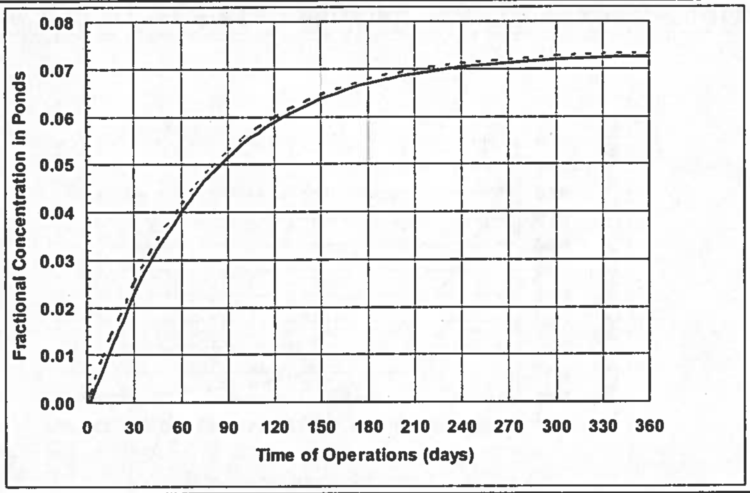
APPENDIX B
EXAMPLES FROM THE NUMI EXPERIENCE

In this appendix, results of calculations based upon the equations derived in the main text and in Appendix A applied to some of the scenarios provided in Table 2 are presented for situations involving NuMI operating conditions as understood as of June 2006. In these calculations, the two values of λ_{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. The condition of the ponds being FULL includes the assumption that the Fox River intake is turned ON while the condition of the ponds being LOW includes the assumption that the Fox River intake is turned OFF. The corresponding values of λ_{source} (FULL) = 1.02 x 10^{-3} day^{-1} and λ_{source} (LOW) = 1.42 x 10^{-3} day^{-1}, corresponding to the two extremes of pond volume used here, are based on a constant discharge of 175 gallons per minute from the NuMI sump. The calculations do not include any tritium that is directly evaporated and does not enter the pond system. For tritium, $\lambda_{tritium}$ = 1.540 x 10^{-4} day^{-1} corresponds to the physical mean-life. The parameter r, unfortunately, has to be estimated. It is now clear upon inspection of data collected during the NuMI operational period and the subsequent shutdown that there is more than one contributor to the tritium concentrations measured in the NuMI sump discharge. In fact, there may be several contributors having individual values of r that vary greatly. It appears that the effective mean-life of the most rapid-clearing component of the concentrations of tritium in the NuMI sump discharge is of the order of 3 days. This corresponds to a value of r of about 0.3 day^{-1} and will be referred to here as the *fast* component. It is now known that there is also a component that is much slower. This *slow* component has a mean-life that has not, as of June 2006, been completely quantified but perhaps is represented by a value of as much as several hundred days, corresponding to an approximate value of r of 0.003 day^{-1}. For each calculation, the sensitivity to this parameter was tested between the range of 3 day^{-1} (clearance mean-life of 0.33 days, far faster than it is known to be) and the "slow" clearance rate of r = 0.003 day^{-1} corresponding to a clearance mean-life of about 330 days. The results for the conditions studied here were all insensitive to values of $r > 0.3$ day^{-1}.

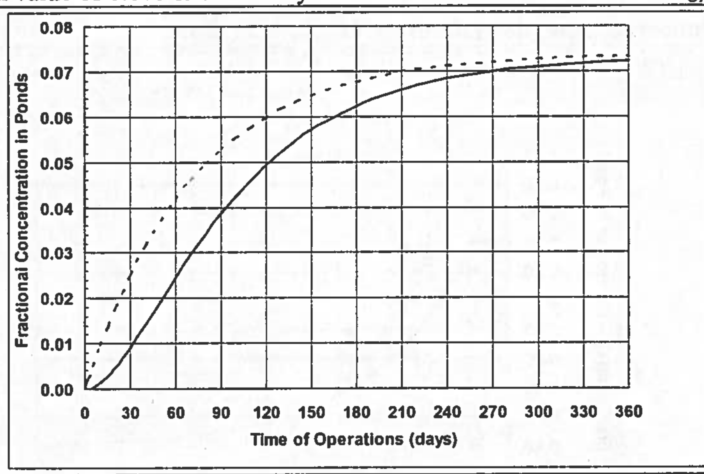
The results of the calculations shown in Figs. 1 - 4 were performed for three values of the parameter r spanning this domain; r = 0.3, r = 0.03, and r = 0.003 day^{-1}. Conditions of both FULL and LOW ponds are considered to set some rough boundaries on the possible results. Figures 1 and 3 calculate the buildup of concentrations of tritium in the ponds during operational periods for the indicated pond system scenario. In each graph for these beam ON scenarios, "Fractional Concentration" is the ratio C_j / s_j. Figs. 2 and 4 calculate the decline of the concentrations of tritium in the ponds during shutdown periods likewise for the indicated pond system scenario. For the beam OFF scenarios, "Fractional Concentration" is measured against the concentration in the ponds at the beginning of the shutdown.

In each plot, the dashed curve is the simplified model of the main text while the solid curve is the more sophisticated model of Appendix A. Only the more sophisticated model is reflective of varying values of r. Thus the dashed curves in all 3 frames of a given panel are the same while the solid curves include the dependence on this variable. In some of the plots, these curves are indistinguishable. A comparison of the two is an indicator of the importance of the simplifying assumptions made in the main text. Since these graphs, essentially of exponential functions, become rather monotonous, and one is mostly interested in the behavior over a period of several months, key parameters for each scenario are provided. For the beam ON scenarios, the equilibrium value reached after an "infinite" irradiation is given along with the characteristic running time, t_{ON}, required to reach 100 x (1 - 1/e) = 63.2 % of that equilibrium value. For the beam OFF scenarios, the characteristic shutdown time, t_{OFF}, required to reach 100 x 1/e = 36.8 % of the initial value of the fractional concentration is provided. The parameters, t_{ON} and t_{OFF}, are thus the effective "time constants" of the processes. These values are those resulting from the use of the more detailed methodology of Appendix A. The presence of multiple delivery mechanisms would require the superposition of individual values.

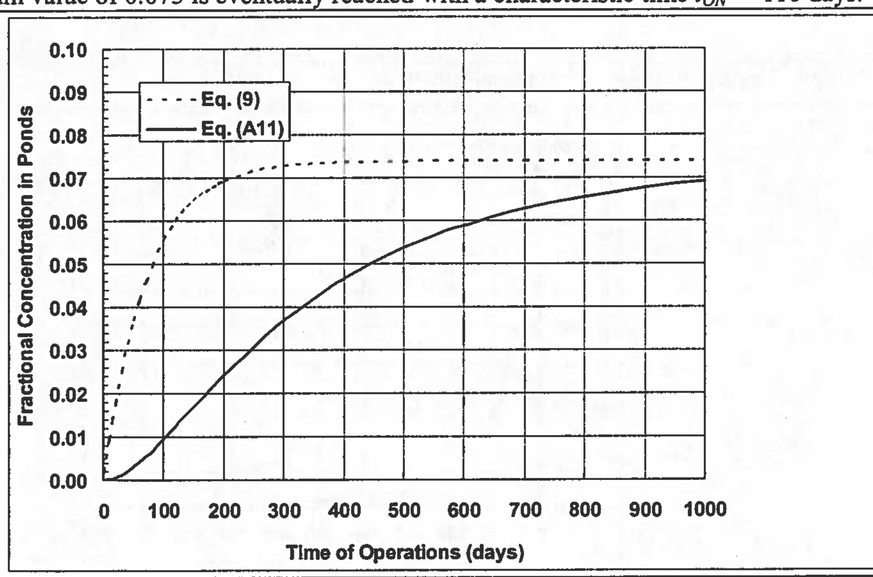
Figure 1 Beam ON, Ponds FULL, Precipitation INTERMEDIATE, Fox River Intake ON



r = 0.3 day^{-1}. An equilibrium value of 0.073 is eventually reached with a characteristic time t_{ON} = 75 days.

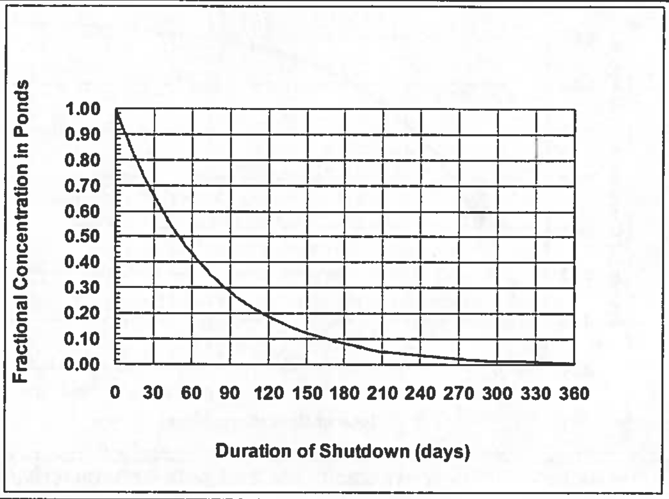


r = 0.03 day^{-1}. An equilibrium value of 0.073 is eventually reached with a characteristic time t_{ON} = 110 days.

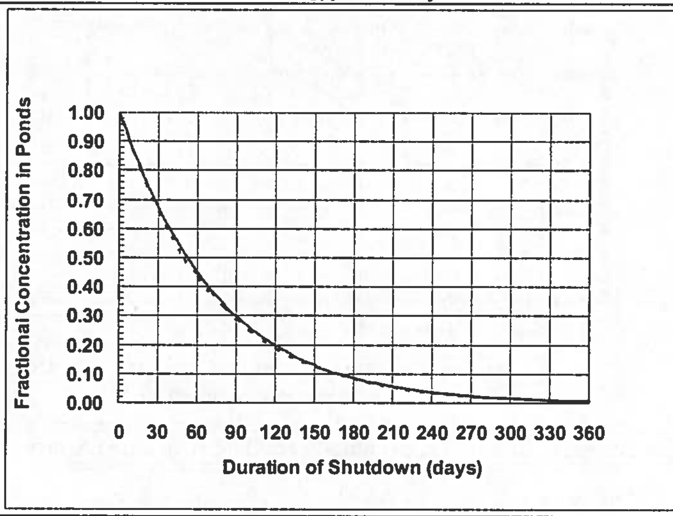


r = 0.003 day^{-1}. An equilibrium value of 0.073 is eventually reached with a characteristic time t_{ON} = 400 days.

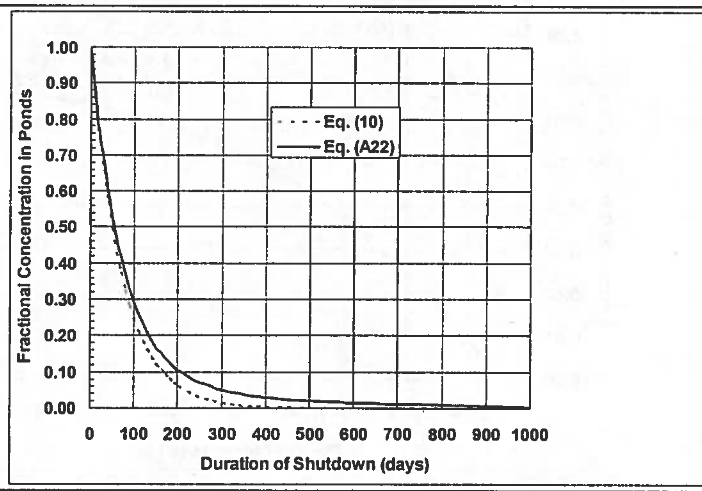
Figure 2 Beam OFF, Ponds FULL, Precipitation INTERMEDIATE, Fox River Intake ON



$r = 0.3$ day^{-1}. The fractional concentration falls to 1/e for $t_{OFF} = 72$ days.

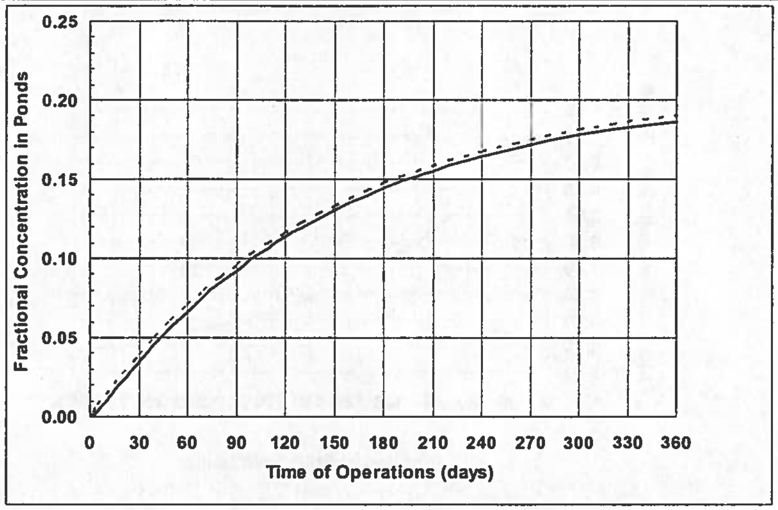


$r = 0.03$ day^{-1}. The fractional concentration falls to 1/e for $t_{OFF} = 75$ days.

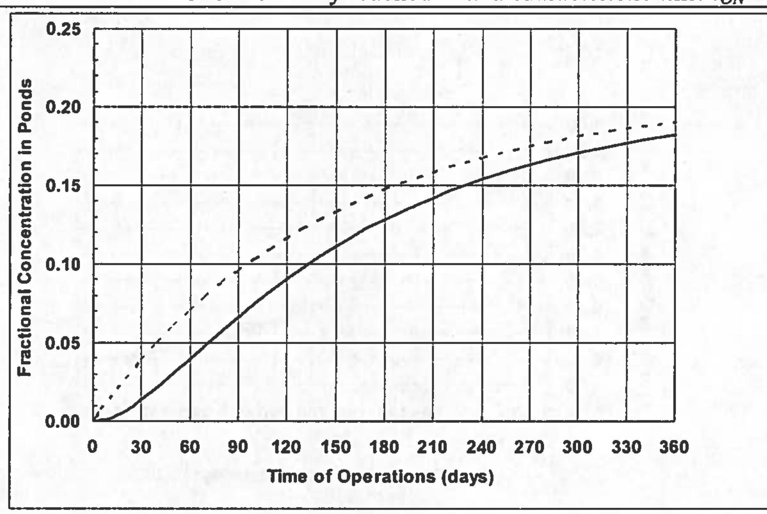


$r = 0.003$ day^{-1}. The fractional concentration falls to 1/e for $t_{OFF} = 80$ days.

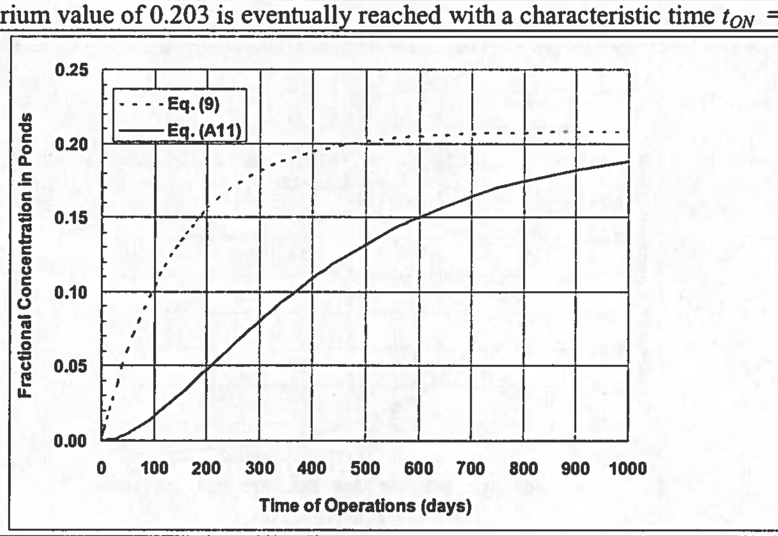
Figure 3 Beam ON, Ponds LOW, Precipitation INTERMEDIATE, Fox River Intake OFF



$r = 0.3$ day^{-1}. An equilibrium value of 0.203 is eventually reached with a characteristic time t_{ON} = 145 days.

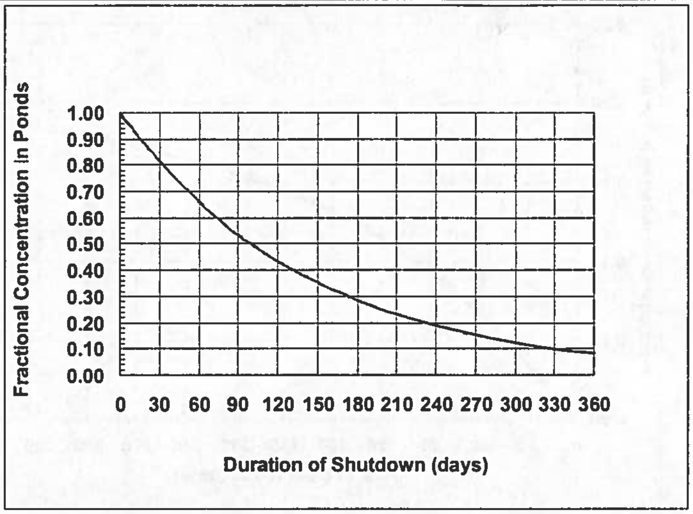


$r = 0.03$ day^{-1}. An equilibrium value of 0.203 is eventually reached with a characteristic time t_{ON} = 180 days.

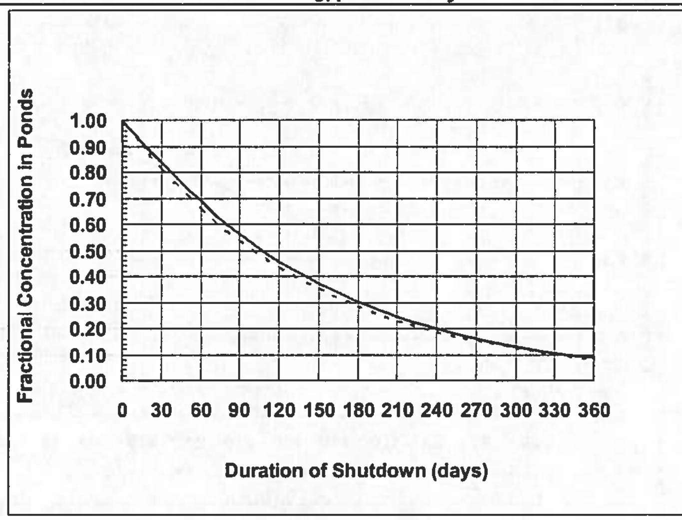


$r = 0.003$ day^{-1}. An equilibrium value of 0.203 is eventually reached with a characteristic time t_{ON} = 480 days.

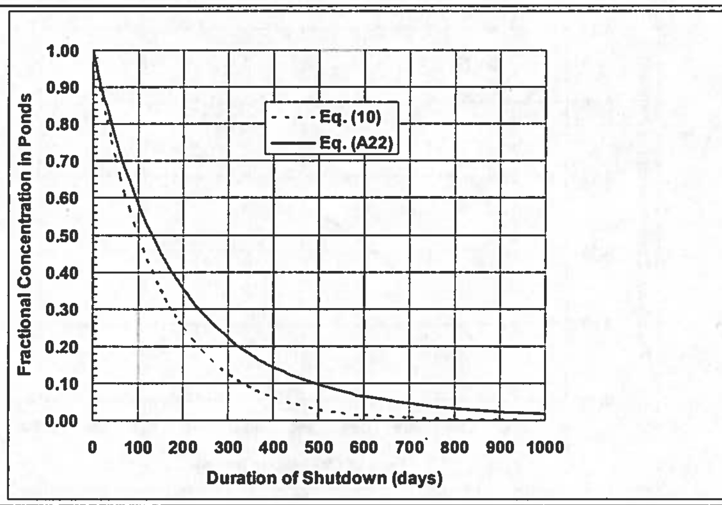
Figure 4 Beam OFF, Ponds LOW, Precipitation INTERMEDIATE, Fox River Intake OFF



$r = 0.3$ day^{-1}. The fractional concentration falls to 1/e for $t_{OFF} = 144$ days.



$r = 0.03$ day^{-1}. The fractional concentration falls to 1/e for $t_{OFF} = 152$ days.



$r = 0.003$ day^{-1}. The fractional concentration falls to 1/e for $t_{OFF} = 192$ days.

A reasonable scenario for approximately the first 120 days of operation of NuMI at high intensity in late summer and autumn of 2005 is likely to be that of "Precipitation INTERMEDIATE, Beam ON, Ponds LOW, and Fox River Intake OFF". During this operational period the concentration in the NuMI/MINOS sump discharge holding tank was perhaps typically 25 pCi cm^{-3}. From the calculations shown in the three panels of Fig. 3, one can get 3 different values of the fractional concentration of tritium in the pond system and crudely estimate the magnitude of the concentrations in the ponds for the corresponding source time constant, r. These are given in Table 3.

Table 3 Estimated Pond Concentration Following 120 Days of NuMI Operations

r (day^{-1})	Fractional Concentration	Estimated Concentration (pCi cm^{-3})
0.3	0.113	2.82
0.03	0.091	2.28
0.003	0.021	0.53

Averaging the results of a site wide sampling of the ponds conducted on November 30, 2005 resulted in an average concentration of 2.4 pCi cm^{-3}. This value is consistent with the above results if the more rapid source component, or components, are dominant.