ENVIRONMENTAL PROTECTION NOTE 10

ANALYSIS OF RADIOLOGICAL EXPOSURE PATHWAYS FOR TRITIUM DISCHARGED TO ON-SITE SURFACE WATERS

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Elaine Marshall and Don Cossairt

Introduction

This paper comprises the technical basis document evaluating known exposure pathways from tritium discharged to the on-site surface waters and also addresses the separate, but related issue, of the releases to the sanitary sewer. It also estimates the radiological impact to a hypothetical member of the public. The calculations included in this document show that the dose equivalent to a such member of the public is very small and current environmental monitoring is sufficient in this regard.

Exposure Pathways

The surface waters on-site are tributaries of much larger water systems, the Fox River and the DuPage River. Both rivers are used for many purposes ranging from irrigation, drinking water and recreational activities (e.g. boating, canoeing, fishing, swimming, and water skiing). As tritium is an isotope of hydrogen, it will be incorporated into the ecological systems in the form of the HTO (tritiated water) molecule. Thus, individuals utilizing the water resources could be expected to receive a dose equivalent from tritium released on the Fermilab site proportionate to the extent the water resources are used and the tritium concentration present.

For example, individuals who consume fish caught in these waters would ingest a quantity of tritium based on the mass of fish ingested over a defined period of time, the equilibrium concentration in the tissue, the preparation method, and storage time. Someone who eats fish caught from these waters three times a week would receive a dose equivalent from this pathway four times greater than the individual who only eats such fish three times a month. Similar conclusions can be made regarding the dose equivalent to an individual based on how much of his drinking water is taken from these waters, the time he spends engaging in recreational activities on or near the water, and the ingested quantities of plant and animal life which utilize this water as their water source.

Another pathway that contributes to dose equivalent from tritium discharged to the on-site surface waters is that of evaporation. Water that evaporates from the surface waters is incorporated into the weather systems and will later become precipitation over a region. It will then be integrated into the ecological systems. Without defining such variables as population density, weather patterns, precipitation, and established ecological systems for that region, it is very difficult to quantify the effect of tritium released on-site on the dose equivalent received by a member of the public through the evaporation pathway.

There are a number of exposure pathways to members of the public from tritium discharged to the surface waters on the Fermilab site. Because of the complexity and the many variables involved, the contribution of each pathway to the dose equivalent received by a member of the public can not be quantified. A worst case scenario was devised that demonstrates the dose equivalent to a member of the public from tritium releases to the surface waters is negligible.

Evaluation of Dose Equivalent to a Member of the Public

The worst case scenario developed assumes that an individual ingests tritium incorporated into drinking water and aquatic foodstuffs (primarily fish). In addition, it postulates that the individual, through recreational uses of the water system, will absorb tritium through his skin. Several assumptions were made in the performance of this study. Those generic to all three of the pathways investigated are stated below. Those pertinent to a specific section are detailed in that section.

- **Fractional uptake into circulation is not affected by the presence of other materials in the water and is equal to 100%. (Reference 1)**
- **Tritium is instantaneously distributed uniformly among all soft tissues. (Reference 1)**
- **The biological half-life of tritium is 12 days. (Reference 2)**

Ingestion of drinking water:

The most significant pathway in which a person could ingest tritium is through drinking water obtained directly from the sump having the highest specific activity. To perform this calculation, in addition to the assumptions stated previously, the following were assumed:

- **The concentration of tritium in the drinking water is 110 pCi/ml.** (Reference 3) The total tritium released off-site for CY 1993 was greater than that released for the previous 4 years. The mean tritium concentration for N01SP4 was the greatest for monitored sumps and the total activity discharged through N01SP4 was the highest of all monitored sumps for CY1993. Since the other sumps have significantly lower concentrations of tritium (5 - 100 times lower) down to levels well below limits of detection, it is asserted that the concentration is much less than this. Any lower concentration of tritium will result in a smaller committed effective dose.
- **The individual drinks the water a rate of 2 liters/day (Reference 4) and this is the individual's only source of drinking water.**

From the concentration and the intake rate, the total activity ingested per year can be calculated.

$$\frac{110\ pCi}{ml} \times \frac{1000\ ml}{liter} \times \frac{1\ \mu Ci}{1E6\ pCi} \times \frac{2\ liters}{day} \times \frac{365\ days}{year} = \frac{80.3\ \mu Ci}{year}$$

For tritium, the Committed Effective Dose Equivalent (CEDE) per unit intake via ingestion is 1.73×10^{-11} Sieverts/Becquerel. (Reference 5) Converting to the more commonly used units, this parameter has the value of 0.064 mrem/µCi.

As the biological half-life of tritium is much less than 50 years, the CEDE for the intake over a year is essentially equal to the dose equivalent received during that particular year and is thus given by:

$$\frac{80.3\ \mu Ci}{year} \times \frac{0.064\ mrem}{\mu Ci} = 5.1\ \frac{mrem}{year}$$

This can be verified by using the DCG values of DOE Order 5400.5. For tritium, the DCG value implies consumption of water with a concentration of 80 pCi/ml will result in a CEDE of 4 mrem/year. Thus, consumption of water with a concentration of 110 pCi/ml will yield a CEDE of:

$$110\ pCi/ml \times \frac{4\ mrem/year}{80\ pCi/ml} = 5.5\ mrem/year$$

which is comparable to the 5.1 mrem/year obtained using the alternative method.

This is considered a highly conservative estimate as the water quality at the sumps is not adequate to encourage drinking. The ditch system combines the discharge of many sumps and rain water. The concentration used here was the highest mean concentration reported for one sump. When the releases from the other sumps and rainfall are combined, the overall concentration would be much lower. In addition, the tritium in the water is further diluted by evaporation and transfer to a much larger water system before being used for drinking water, yielding an even lower committed effective dose equivalent.

Consumption of Fish:

Another pathway worth consideration is the ingestion of tritium contained in the tissues of aquatic animals, primarily fish. The plant and animal life exposed to tritiated water would incorporate the tritium into its tissue. To perform this calculation, in addition to the general assumptions made previously, the following were assumed:

- **The fish are assumed to inhabit an aquatic environment with a tritium concentration of 11.9 pCi/ml (See Appendix for the calculation of this particular postulated specific activity.) and to have a concentration of tritium equal to 11.9 pCi/g.** This value is derived from assuming a density of tissue of 1.0 g/ml and that the tissue has a concentration equal to that of the environment. It is likely that the fish and other organic matter would have a lesser concentration. It is expected that the tritium concentration in the ponds, where fish of significant size could be found, to be much lower than this value. With a lower concentration of tritium in the aquatic environment, the fish tissue would attain a lower equilibrium value. Thus, lower concentrations of tritium in the aquatic environment will result in a smaller committed effective dose.

- **An individual consumes 1/2 pound of fish (0.227 kg) per meal, three times per month.**

From the concentration and the intake rate, the total activity ingested per year can be calculated which can then be used to determine the dose equivalent for the year.

$$\frac{0.227\ kg}{meal} \times \frac{3\ meals}{month} \times \frac{12\ month}{year} = 8.17\ kg\ tissue/year$$

$$\frac{8.17\ kg\ tissue}{year} \times \frac{1000\ g}{kg} \times \frac{11.9\ pCi}{g} \times \frac{\mu Ci}{1E6\ pCi} = 9.72E-2\ \mu Ci$$

$$\frac{9.72E-2\ \mu Ci}{year} \times \frac{0.064\ mrem}{\mu Ci} = 6.2E-3\ \frac{mrem}{year}$$

As was the case with the ingestion of drinking water, this is considered to be a very conservative estimate. It does not take into account any decay of the tritium from the time of capture to the time of ingestion. It also does not take into account any effect method of preparation may have on the concentration of tritium. Both of these would serve to decrease the concentration of tritium in the tissue. Lower concentrations of tritium in the tissue would yield a lower committed effective dose equivalent.

Recreational Activities:

In considering recreational activities, the significance of the pathway in which tritium would be absorbed through the skin can be determined. Several assumptions were made for this calculation in addition to the general assumptions made previously.

- **The individual would be swimming in Casey's Pond where the concentration is equal to 11.9 pCi/ml. (See Appendix for the calculation of this particular postulated specific activity.)** Swimming and other such recreational activities are not permitted on the Fermilab site. Given the dilution known to occurred prior to an off-site recreational activities, the dose equivalent to a member of the public would be significantly lower.

- **Uptake from body of water with a concentration of 11.9 pCi/ml through the skin is 0.119 pCi/min.** No information was available for absorption through the skin for complete submersion in tritiated water. This was assumed to be similar to the uptake for absorption through the skin by immersion in a cloud of tritium vapor. (Reference 1) A decrease in the rate of uptake would result in a lower committed effective dose equivalent.

- **The individual swims or participates in other recreational activities for 3 hours/week during the months of June, July, August and September.** The water and air temperatures during other months of the year in this climate prohibit these activities. Any decrease in the amount of time spent participating in such activities would serve to decrease the committed effective dose equivalent received.

From the rate of uptake, the total activity absorbed through the skin per year can be calculated which can then be used to determine the dose equivalent for the year.

$$\frac{0.119\,pCi}{min} \times \frac{\mu Ci}{1E6\,pCi} \times \frac{60\,min}{hr} \times \frac{3\,hr}{week} \times \frac{17\,week}{year} = 3.64E-4\,\frac{\mu Ci}{year}$$

$$\frac{3.64E-4\,\mu Ci}{year} \times \frac{0.064\,mrem}{\mu Ci} = 2.3E-5\,\frac{mrem}{year}$$

Total Effective Dose Equivalent from Tritium Released to the On-Site Waterways

The total effective dose equivalent estimated in this worst case scenario is only a small fraction of the 360 mrem/year (Reference 6) that an individual would receive from natural background, medical sources, and consumer products. In addition, this value is well below the 100 mrem per annum dose equivalent allowed to members of the public by the DOE Order 5400.5. Given the controls in place, the unlikelihood of this scenario, the levels of activity actually measured, and the large dilution that occurs before a member of the public would be drinking this water or using it for recreational purposes, the actual dose equivalent to a member of the public from discharge of tritium to the surface waters on the Fermilab site is negligible. Additional analysis of the various exposure pathways and environmental monitoring would provide no benefit.

Tritium in the Regeneration Effluent

On May 20, 1994, Fermilab made its first regeneration effluent discharge to the Batavia Sanitary Sewer System. A total of 46 discharges were made between May 1994 and December 1994 for a total discharge volume of 147,908 gallons. The total activity released to the sanitary sewer was 0.442 Ci for an average concentration of 0.790 pCi/ml. (Reference 7) The activity concentrations were measured immediately prior to discharge.

Fermilab, at the lower end of the range, releases 70,000 gallons per day to the Batavia Sanitary Sewer System (Reference 8) or 2.56E7 gallons per year. Scaling the releases of the regeneration effluent to date through the entire year, the volume of effluent over a typical year can be estimated.

$$147908\,\frac{gallons}{year}\times\frac{365\ days}{225\ days}=2.40E5\,\frac{gallons}{year}$$

Diluting the regeneration effluent by the total release to the sewer, yields a dilution factor equal to:

$$\frac{2.40E5\ gallons/year}{2.56E7\ gallons/year}=9.39E-3$$

Therefore, the average concentration released from the site is 7.42E-3 pCi/ml. Assuming an individual drinks 2 liters of the effluent per day for 365 days of the year and has no other source of drinking water, the total quantity of tritium ingested can be calculated which can then be used to determine the dose equivalent rate.

$$\frac{7.42E-3\ pCi}{ml}\times\frac{1000\ ml}{liter}\times\frac{1\ \mu Ci}{1E6\ pCi}\times\frac{2\ liters}{day}\times\frac{365\ days}{year}=\frac{5.42E-3\ \mu Ci}{year}$$

$$\frac{5.42E-3\ \mu Ci}{year}\times\frac{0.064\ mrem}{\mu Ci}=3.5E-4\,\frac{mrem}{year}$$

If the dilution from water treatment were considered, the dose equivalent rate would be much lower. The Batavia Waste Water Treatment Plant has an average capacity of 2.5 - 3 million gallons per day (Reference 9) or 9.125E8 gallons per year. This further dilutes the regeneration effluent by a factor of:

$$\frac{2.40E5\ gallons/year}{9.13E8\ gallons/year} = 2.63E-4$$

Again, assuming an individual drinks 2 liters of the water per day for 365 days of the year and has no other source of drinking water, the total quantity of tritium ingested can be calculated which can then be used to determine the dose equivalent rate.

$$\frac{2.08E-4\ pCi}{ml} \times \frac{1000\ ml}{liter} \times \frac{1\ \mu Ci}{1E6\ pCi} \times \frac{2\ liters}{day} \times \frac{365\ days}{year} = \frac{1.52E-4\ \mu Ci}{year}$$

$$\frac{1.52E-4\ \mu Ci}{year} \times \frac{0.064\ mrem}{\mu Ci} = 9.70E-6\ \frac{mrem}{year}$$

If the individual's drinking water was supplemented by other sources the amount of tritium would be decreased even more. Thus, the dose equivalent that would be received by any member of the public from this source is negligible.

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APPENDIX

Concentration in Casey's Pond

Casey's Pond has a constant volume V (1.38E11 cm^3) representing a balance between inflow, outflow, and evaporation.

Step 1: Estimate the concentration of the input into Casey's Pond.

Assuming that the entire flow rate into Casey's Pond is from the sumps, the sumps all have an equal flow rate, and the concentrations measured at the monitored sumps are representative of all sumps feeding into Casey's Pond, an activity concentration can be estimated for the flow into the pond. Monitoring is performed currently at suspect sumps, biasing the overall concentration derived for this study in the upward direction. In addition, a significant fraction of the volume into Casey's Pond originates as rain water which dilutes the input from the sumps. Thus, actual concentrations in Casey's Pond would be much lower than this estimation.

Fifteen sumps were monitored for tritium during CY1993. (Reference 3) Assuming that all the sumps contribute equally to the concentration, the average concentration C into Casey's Pond is 12.5 pCi/ml.

Step 2: Calculate the number of atoms of H-3 which enter the pond per unit time. Assuming the inflow maintains a constant concentration C (pCi/ml) and constant flow rate.

$$Inflow\ (\frac{atoms}{s}) = \frac{C}{\lambda} v$$

where λ = decay constant of H-3 (sec^{-1})
v = flow rate into Casey's Pond ($cm^3\ sec^{-1}$)

Step 3: Determine the total number of H-3 atoms in the pond at time t.

$$\frac{dN(t)}{dt} = Inflow - Decay - Outflow - Evaporation \& Other\ Losses$$

$$\frac{dN(t)}{dt} = \frac{C}{\lambda} v - \lambda N(t) - \frac{(D+E)}{V} N(t)$$

where N(t)=total number of atoms at time t
D=discharge rate from the pond ($cm^3\ sec^{-1}$)
E=loss rate from the pond ($cm^3\ sec^{-1}$)

Solving for N(t):

$$N(t) = \frac{\frac{Cv}{\lambda}}{\lambda + \frac{(D+E)}{V}}\left[1 - \exp\left(-\lambda t - \frac{(D+E)t}{V}\right)\right]$$

At time t = 0, the number of tritium atoms in the pond, N(0) = 0. After a time comparable to the operational period of the laboratory, a steady state will have been achieved and t can be taken to be infinity.

$$N(\infty) = \frac{\frac{Cv}{\lambda}}{\lambda + \frac{(D+E)}{V}} = \frac{\frac{VCv}{\lambda}}{\lambda V + D + E}$$

Thus, the activity concentration in the pond at t = ∞, in pCi/ml, is:

$$\frac{A(\infty)}{V} = \frac{\lambda N(\infty)}{V} = \frac{Cv}{\lambda V + D + E}$$

(Following taken from Reference 10) The flow rate into Casey's Pond is dependent upon the operational mode and the season of the year. During Fixed Target Operation, the minimum flow rate is 6600 GPM and the maximum flow rate is 10,300 GPM. At other times, the minimum flow rate is 2600 GPM and the maximum is 4600 GPM. Whatever the flow rate into Casey's Pond, an equivalent amount is removed for ICW Supply. Thus, v = D = 10,300 GPM = 6.50E5 cm^3s^{-1}. The make-up water, to account for losses due to evaporation and drainage, ranges from 500 GPM to 1000 GPM. Worst case scenario would take E = 500 GPM = 3.15E4 cm^3s^{-1}.

$$Activity\ Concentration\left(\frac{pCi}{ml}\right) = \frac{\left(12.5\frac{pCi}{cm^3}\right)\left(6.498E5\frac{cm^3}{s}\right)}{\left(1.787E-9s^{-1}\right)\left(1.378E11\,cm^3\right) + \left(6.498E5\frac{cm^3}{s}\right) + \left(3.154E4\frac{cm^3}{s}\right)}$$

$$Activity\ Concentration\left(\frac{pCi}{ml}\right) = 11.9\frac{pCi}{ml}$$

The activity concentration for the water in Casey's Pond is truly a worst case estimate based upon data for the monitored discharges from suspect sumps. Suspect sumps are only those which have a potential for containing radioactivity and make up approximately 10% of the sumps. Rain water was neglected, but could easily account for 20-30% of the total inflow volume. The extreme flow rates were assumed. However, the maximum inflow would occur during the summer, corresponding to the maximum input of make-up water, not the minimum assumed for this model. Measured values to date of samples taken from Casey's pond have been below detection limits (typically 0.1 pCi/ml) and provide evidence of the actual lower concentrations.