



Fermilab
 ES&H Section

ASSESSMENT OF OUTDOOR STORAGE OF RADIOACTIVE MATERIALS

David Boehnlein, Vernon Cupps, Kathy Graden and Sharon Templeton

October, 1996

Written By: David Boehnlein Date: 1/17/97
 David Boehnlein

Written By: Vernon P. Cupps II Date: 1/16/97
 Vernon Cupps

Written By: Kathy Graden Date: 1/20/97
 Kathy Graden

Written By: Summer employee, not available Date: _____
 Sharon Templeton

Reviewed By: Don Cossairt Date: 1/27/97
 Don Cossairt, RP Group Associate Head

Reviewed By: Rod Walton Date: 1/22/97
 Rod Walton, EP Group Associate Head

Approved By: William J. Griffing Date: 1/27/97
 Bill Griffing, ES&H Section Head

Distribution via QuickMail (approved version on file):
 R.P. Staff, EP Staff, D. Cossairt

ASSESSMENT OF OUTDOOR STORAGE OF RADIOACTIVE MATERIALS

David Boehnlein, Vernon Cupps, Kathy Graden and Sharon Templeton

October, 1996

INTRODUCTION

The Railhead area on the Fermilab site is used for the storage of known and potentially radioactive materials. These materials are items which are or may have been activated by particle beams due to operations at Fermilab. A relatively large amount of low-level radioactive material (approximately 50,000 cubic feet with dose rates less than 1 mrem/hour) in the form of metal system components is stored on hardstand at the Railhead for potential reuse. Since these materials are exposed to ambient weather conditions, often for extended periods of time, the question arises as to whether corrosion might pose a potential contamination concern. Previous measurements^{1,2} have not indicated any such problems. However, a comprehensive study focused on potential environmental concerns was prompted by observations of the Tiger Team which visited Fermilab in 1992.³ This note describes the results of that study.

SCOPE

This section describes a survey of the materials stored at the Railhead and a set of conservative assumptions which are used to estimate a source term for the deposition of radioactivity there.

In October, 1994, items at the Railhead were surveyed to estimate typical dose rates of radioactive materials stored outdoors.⁴ It was assumed that all items at the railhead are radioactive to some extent. The background rate at the railhead is too high to determine whether an item exceeds Fermilab's release criteria for radioactivity. The following estimates are made from both the data collected during the railhead survey and the SMR data printout provided by Frank Beverley of the Business Services Section. Dimensions of each item are taken from the SMR data printout.

1. At that time there were 1649 items in the railhead inventory.
2. The following estimates were made from the survey:

Dose rate (mR/hr on contact) Percent of all items inventory

Less than 0.1	35.6 %
0.1 - 0.9	60 %
1.0 - 4.9	3 %
5 - 9.9	0.8 %
10 and greater	0.6 %

3. It is estimated that about 40% of the items are painted. We cannot determine if the items are painted before or after being radioactivated.
4. The following is a breakdown of the types of materials in the railhead inventory. The percentages are based on the total amount of surface area exposed to weathering:

Steel	90.6 %
Copper	8.9 %
Aluminum	0.4 %
Other (mostly concrete)	0.1 %

5. The survey data used in the subsequent calculations comprise roughly 8% of the total railhead inventory. The items surveyed are assumed to be a representative sample.

SAMPLING PROTOCOL

Sample Grid Determination

A number of samples were collected from the Railhead to determine whether corrosion from these items is contaminating the area or the environment with radioactivity. These samples were analyzed in Fermilab's Activation Analysis Laboratory (AAL) both for tritium and accelerator-produced gamma-emitting radionuclides. Copies of the AAL reports are included in Appendix 1.

One sample of run-off water from the southwest corner of the Railhead was analyzed. This sample was collected from standing water in a field just beyond the Railhead fence and at slightly lower ground elevation. The sample was taken shortly after a heavy rain. The rationale for choosing this location was that the run-off water would be the most likely medium for the prompt transfer of radioactivity to the environment, if any such transfer were taking place. No significant radioactivity was detected in this sample.

The next step taken to address potential contamination was determining those areas of highest potential contamination based on the level of radioactivity of stored materials and the length of storage time. Discussions with Kevin Orcutt of the Business Services Section revealed that activated materials were stored in the southern half of the Railhead and that three areas were considered to have the highest potential for contamination. Area I is near the east end of Row 6 along the north eastern fence line between the green magnets to the north and the metal plates to the south (see Figure 1). Area II is inside Nevis Barn at the southeastern end. Area III is located where radiation area magnets were stored behind Nevis Barn along the south side fence.

The second step was to determine whether a valid approach for this study was to utilize the IEPA grid interval equation to calculate a statistically valid sampling grid and thereby dictate the number of samples required to demonstrate clean closure. The IEPA grid interval equation is as follows:

$$GI = \frac{1}{2} \sqrt{\frac{A}{\pi}} \quad (1),$$

where GI is the grid interval measured in feet and A is the area to be sampled in square feet. Initial calculations using square footage of each defined area resulted in the grid interval values and number of required samples as shown in Table 1. The number of samples to be collected in each of the three areas (18, 14 and 21 respectively) was considered to be too cumbersome for the purpose of this study. Thus another calculation was performed, this time, on the whole southern area that has at times been used for activated materials storage. The calculated grid interval for the larger area resulted in the requirement for 17 samples. Seventeen samples was also considered to be cumbersome at this point in the study particularly considering the expected low level concentrations of

radionuclides in the gravel/soil and the time and cost to analyze this number of samples. It became clear at this point that a more cost-effective procedure was necessary.

Table 1. Grid Interval Calculation Parameters

	<u>Area I (Row 6)</u>	<u>Area II (Nevis Barn)</u>	<u>Area III (behind Nevis Barn)</u>	<u>Southern Area (total)</u>
length	60 ft	120 ft	30 ft	odd shape
wide	30 ft	32 ft	10 ft	odd shape
area	1800 sq ft	3840 sq ft	300 sq ft	175234 sq ft
grid interval	12	17.5	4.9	118.1
# of samples	18	14	21	17

Sample Collection

An alternative procedure was adopted which focused on first sampling areas where the highest concentrations of radionuclides could reasonably be expected to exist. If no concentrations which would be cause for concern were found there, they could not plausibly be found anywhere at the Railhead. If such concentrations were found, then a more extensive sampling plan could be considered. The criteria which we would consider "cause for concern" are discussed in the section on Potential for Contamination Problems below.

Samples were collected at depths of approximately 2 inches and 12 inches. Figure 1 is a map showing the locations of Railhead samples. The results of analyzing these samples are summarized in Table 2.

A set of background samples was collected to ascertain how much, if any, of the radionuclide content in the Railhead soil samples might be attributable to the materials stored there. Figure 2 shows the location of site wide background samples. The site wide samples include three surface soil samples and two surface gravel samples. The three surface soil sample locations were selected from relatively mature tree stands that have likely been undisturbed since the commencement of activities at Fermilab. The two gravel samples were selected from an area of low traffic (near farm site 5), and an area with higher non-laboratory related traffic (Anthony Frelow flying field parking lot). These sample locations were selected to provide information on background radionuclide concentrations in natural soils and in imported gravels used for hardstand construction.

Based on the results shown in Table 2, additional sampling was deemed necessary at the 'hottest' location at the east end of Row 6. Deeper samples were collected to provided a vertical profile of radionuclide and tritium concentration with depth. Assistance from the roads department was required to temporarily remove the hardstand at this location so that the samples could be collected from natural soil at depths of 20 and 40 inches.

Sample Collection Procedures

The following procedures were used during the sampling of water, gravel and soil samples.

- 1) Selected exact sample location within the targeted areas.
- 2) For grab gravel/soil samples:
Carefully cleared debris off the ground surface. Collected a gravel/soil sample at a depth of approximately three inches and another at a depth of 12 inches using a

shovel or trowel. Followed procedure 4) between samples. Deposited each sample in a separate sample bottle.

For profile samples:

Dug a five foot pit utilizing a back hoe. Collected samples at depths of 20 inches and 40 inches. Followed procedure 4) between samples. Deposited each sample into its own individual sample bottle. Measured and documented sample depths. Filled pit.

For surface water samples:

Used a "dipper" attached to a pole to obtain an undisturbed sample. Poured sample into a sample bottle.

3) Wiped the sample bottle clean with a kim-wipe and labeled the bottle with a Sample Identification Number. Documented sample in the field note book and on the Chain of Custody Sheet. Placed clean sample bottle in a plastic zip lock bag ensuring that the sample identification could be clearly seen through the bag.

4) Cleaned all sample equipment in the wash basin with Alconox and tap water. Rinsed equipment two times with distilled water over the wash basin. Emptied wash basin into a 55 gallon drum.

5) Repeated procedures 1 through 4 until all samples had been collected.

6) Transported samples to 21 Shabbona and store in the locked (1A-7 key) environmental sample refrigerator. Made a copy of the Chain of Custody form, and label it COPY 1. Placed the original Chain of Custody form in the plastic jacket on the outside of the refrigerator door. Filled out an AAL Work Request form and placed in the request box. Placed COPY 1 in the current Railhead Soil Survey file, Environmental Protection Group office on WH7E. The original Chain of Custody will remain with the samples until the final analytical results have been obtained and verified, and sample disposal has been completed. Following disposal of the samples, place the original Chain of Custody in the Railhead Soil Survey file with COPY 1.

Analytical Results

Analytical results of the surface water, gravel and soil samples are contained in Table 2. The surface water sample, located at the southwestern corner of the Railhead, is below the DOE 5400.5 ingested water Derived Concentration Guide (DCG) limits for ^{137}Cs , ^{60}Co , ^{40}K and ^3H . The measured concentrations are well within these limits.

The gravel and soil sample results are not so simple. The implications of the results are discussed in detail in the next section. Measurable amounts of ^3H are found in the Row 6 sample @ 12" and the sample behind Nevis Barn @ 2".

The gravel and soil samples both on the Railhead site and at the background locations around Fermilab all indicate varying concentrations of ^{40}K .

Two samples contain ^{60}Co . Both are shallow 2 inch deep samples at the east end of Row 6 and behind Nevis Barn.

Samples from the east end of Row 6 at depths of 2 inches and 12 inches and three background soil samples all contain ^{137}Cs .

Table 2. Radionuclide and Tritium Results for Surface Water , Gravel and Soil Samples

<u>Location</u>	<u>Matrix</u>	<u>^{137}Cs (pCi/g)</u>	<u>^{60}Co (pCi/g)</u>	<u>^{40}K (pCi/g)</u>	<u>^3H (pCi/g)</u>
DOE 5400.5		3	10	7	2000
SW Corner Railhead	surface water	nd	nd	nd	<1.0
E end Row 6 @ 2"	gravel	0.05	0.14	3.13	<1.0
E end Row 6 @ 12 "	soil	0.22	nd	11.3	1.3
E end Row 6 @ 20"	soil	nd	nd	13.5	
E end Row 6 @ 40"	soil	nd	nd	13.7	
Nevis Barn @ 2"	gravel	nd	nd	4.26	<1.0
Nevis Barn @ 12"	gravel	nd	nd	6.34	<1.0
Behind N. Barn @ 2"	gravel	nd	0.22	4.58	3.3
Behind N. Barn @ 20"	gravel	nd	nd	6.50	<1.0
S end Swenson Rd	gravel	nd	nd	1.99	<1.0
Anthony Frelow parking lot	gravel	nd	nd	3.51	<1.0
Farm site 74 - trees	soil	0.36	nd	16.3	<1.0
N side Pine St. entrance - trees	soil	0.63	nd	9.73	<1.0
Farm site 14 - trees	soil	0.70	nd	13.4	<1.0

Additional Information

Additional information including background notes, grid interval calculations, maps and field notes are contained in a file submitted to Paul Kesich. Table 3 contains a cross reference list of sample numbers and descriptive sample locations.

Table 3. Sample Cross Reference

<u>Location</u>	<u>Sample Number</u>
SW Corner Railhead	960531KO01
E end Row 6 @ 2"	960619ST01
E end Row 6 @ 12 "	960619ST02
E end Row 6 @ 20"	960909ST01
E end Row 6 @ 40"	960909ST02
Nevis Barn @ 2"	960619ST03
Nevis Barn @ 12"	960619ST04
Behind N. Barn @ 2"	960619ST05
Behind N. Barn @ 20"	960619ST06
S end Swenson Rd	960820ST01
Anthony Frelow parking lot	960820ST02
Farm site 74 - trees	960820ST03
N side Pine St. entrance - trees	960820ST04
Farm site 14 - trees	960820ST05

Interpretation of Analytical Results

Cesium-137 is not found as an activation product in the materials stored at the Railhead. It is a fission product which was produced in considerable quantities in the above-ground nuclear tests conducted in the 1940's and '50's. Since ^{137}Cs has a 30 year half-life, it is still commonly found in fallout today. Although Fermilab possesses small quantities of

^{137}Cs in the form of encapsulated sources, these are rigorously controlled and there is no plausible means of finding the observed distribution of ^{137}Cs other than by means of fallout. Hence, no further consideration is given to this radionuclide here.

Potassium-40 is found in the naturally-occurring background radioactivity. Its ubiquitous appearance among the samples, both those at the Railhead and those taken for background studies, clearly indicate that the materials stored at the Railhead are not the source of ^{40}K . The presence of ^{40}K as background radioactivity in the local soil confirms the findings of studies performed early in Fermilab's history.⁵ Although these studies also found ^3H in the soil's radioactive background, the samples seem to indicate elevated levels at some locations in the Railhead area, so this radionuclide is considered in the next section.

Potential for Environmental Contamination

Two potential contamination problems are considered here: 1) surface contamination and the potential for spreading it or for persons being exposed to it; 2) contamination of the environment via runoff water or seepage of radioactivity into groundwater.

The first consideration is easily addressed. The samples, supported by wipe tests and radiation surveys, clearly show that there is no level of contamination present which could plausibly be considered a hazard to personnel. Although such surveys are not sensitive to tritium, it is not plausible that significant amounts of accelerator-produced tritium would be present in the absence of all other accelerator-produced radionuclides, which such surveys would readily detect. Chapter 2 of the Fermilab Radiological Control Manual specifies the levels of removable radioactivity which would cause an area to be posted as a Contamination Area and the levels of ground contamination fall well below those limits. There is no plausible way for a worker to spread contamination after working in the Railhead area. The levels are simply too low.

Environmental concerns require more detailed consideration. There is no regulatory guideline to indicate what is an acceptable level of radioactivity in soil. The only numbers specified by DOE are the DCGs, which apply to water. In order to apply them to this study, some assumptions must be made about how radioactivity might find its way into the water. Two mechanisms are considered: direct runoff through rainwater and seepage through soil to a ground aquifer. The radionuclides ^3H and ^{60}Co are considered separately.

Tritium

Tritium is a highly mobile radionuclide, yet was found in high concentration in only one spot near the surface. It was not detected at a high concentration deeper in the soil nor was the runoff water observed to have a high concentration of tritium. If, in fact, the tritium is being leached from the Nevis blocks, it seems to be greatly diluted by the time it is transported very far from that point and hence does not threaten to raise the level of radioactivity to DCG levels either in runoff or in the aquifer. Additional sampling in the vicinity of the Nevis blocks might be advisable to verify this.

^{60}Co

Cobalt-60, on the other hand, is evidently less mobile than tritium. The fact that it was found in the vicinity where metal items were once stored but ^{54}Mn was not found there may indicate that the ^{60}Co had been there for a number of years, since both radionuclides would be expected to occur in activated metals such as copper or brass. Cobalt-60 has a half-life of approximately five years, while ^{54}Mn has a half life of approximately one

year. If the radioactivity was deposited there only four years or so ago, the ^{54}Mn might well have decayed away to undetectable levels, while much of the ^{60}Co remained. In this time, however, the ^{60}Co had not migrated to a depth of 20". One might realistically assume a depth migration of only a few inches per half-life, which would essentially cause the ^{60}Co to disappear long before it reached the aquifer. Furthermore, no ^{60}Co at all was seen in the runoff sample. Hence it is reasonable to conclude that the ^{60}Co at the Railhead does not present an environmental hazard.

These suppositions are borne out by the work of Leddicotte *et. al.*⁶ In their studies of potential environmental hazards due to the shallow burial of low-level radioactive waste, they determined allowable concentration limits in soil based on the potential for radionuclide migration into groundwater. These limits were taken to be 0.01 MPC. The MPC is the Maximum Permissible Concentration of the radionuclide, a quantity which has since been replaced by the Derived Concentration Guide. The MPC for ^{60}Co was 5×10^{-5} $\mu\text{Ci/ml}$ (DCG = 1×10^{-5}) and the MPC for tritium was 2×10^{-3} $\mu\text{Ci/gm}$ (same as DCG). For the purposes of this note, the difference can be ignored. The suggested limit for ^{60}Co was essentially infinite ($> 9 \times 10^{99}$ nCi/gm), indicating that the rate of migration is so slow that the radioactivity would have entirely decayed before reaching the perched water table.

Leddicotte *et. al.* have also suggested a limit for the concentration of tritium, again based on considerations of site release to ground water. The suggested limit for tritium is 5×10^5 nCi/gm. Since the soil density is taken to be 2.5 gm/cm^3 , this implies a limit of approximately 1.3×10^6 nCi/ml. This number is many orders of magnitude larger than any observed anywhere on the Fermilab site.

Similar work has been done by Staley *et. al.*⁷ This group modeled the migration of radionuclides from buried radioactive waste, including structural materials from a decommissioned nuclear reactor. We consider here only that part of the study pertaining to the structural materials, since they are most similar in nature to the items in storage at the Railhead, although other forms of waste were also modeled. A ^{60}Co source term of 470 Ci was assumed (for the structural components only). The maximum concentration in groundwater at a distance of 100 m was modeled for several different types of soil. For clay and silt, there was no significant amount of ^{60}Co . The source term used makes this a very reasonable calculation when applied to the Railhead: $470 \text{ Ci}/1649 \text{ items} = 285 \text{ mCi/item}$ on average. Using calculations performed by Cossairt⁸ for activation of a steel magnet, a dose rate of 1 mrad/hour corresponds to a total activity of 0.27 Ci for a typical main ring magnet. Although higher dose rates are observed for some items, many of the items at the Railhead are considerably smaller than a main ring magnet.

These theoretical calculations are supported by the experimental work in reference 5. In these studies, soil samples from the Fermilab site were irradiated using particle beams, inducing radioactivity in them. The induced activity was measured using γ -ray spectroscopy. Water was then leached from the samples and its radioactivity content was measured. Although ^{60}Co was observed in the activated samples, it was not found in the leached water.

CONCLUSION

Although the outdoor storage of radioactive material does not pose either a significant occupational or environmental hazard while Fermilab is in operation, it is necessary to

consider actions to be taken during the Decontamination and Decommissioning of Fermilab, when such time comes. At that time, further sampling will be necessary and the results will have to be compared with whatever cleanup standard is in effect at that time. It is possible that some remedial action, such as soil removal will have to be taken before the area could be released for public use. The authors are indebted to Steve Benesch for analysis of the samples taken and to Kevin Orcutt for his assistance in the selection of sampling sites.

¹ J. D. Cossairt and M. K. Oliver, *Surface Contamination Due to Stored Radioactive Steel and Concrete*, Fermilab Radiation Physics Note 34 (1982).

² S. Bluma, Memo to C. Zonick, Subject: Railhead Radioactive Material Storage Area, Wipe Survey (January 19, 1994).

³ Fermilab Tiger Team Corrective Action Plan (1992).

⁴ K. Graden and K. Orcutt, personal communication.

⁵ T. B. Borak, M. Awschalom, W. Fairman, F. Iwami and J. Sedlet, *The Underground Migration of Radionuclides Produced in Soil Near High Energy Proton Accelerators*, *Health Physics*, **23**, 679 (1972).

⁶ G. Leddicotte, W. Rodger, R. Frenberg, and H Morton, *Suggested Quantity and Concentration Limits to be Applied to Key Isotopes in Shallow Land Burial*, Management of Low-Level Radioactive Waste, M. Carter, A. Moghissi, and B. Kahn, ed.; Pergamon Press, New York (1979).

⁷ G. Staley, G. Turi, and D. Schreiber, *Radionuclide Migration from Low-Level Waste: A Generic Overview*, Management of Low-Level Radioactive Waste, M. Carter, A. Moghissi, and B. Kahn, ed.; Pergamon Press, New York (1979).

⁸ J. D. Cossairt, *Magnet Activation Calculation*, Fermilab Radiation Physics Note #22 (May 1979).