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Evaluation of the Radioactive Inventory in, and Estimation of Isotopic Release From, the Waste in Eight Trenches at the Sheffield Low-Level Waste Burial Site

Prepared by D. R. MacKenzie, J. F. Smalley, C. R. Kempf, R. E. Barletta

Brookhaven National Laboratory

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D. R. MacKenzie, J. F. Smalley, C. R. Kempf, R. E. Barletta

Brookhaven National Laboratory
Upton, NY 11973

Prepared for
Division of Waste Management
Office of Nuclear Material Safety and Safeguards
U.S. Nuclear Regulatory Commission
Washington, D.C. 20555
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ABSTRACT

An inventory has been compiled of the isotopes of half-life >5 years buried in eight of the trenches at the Sheffield low-level waste burial site. This was accomplished through a search of the Sheffield radioactive shipment records (RSRs). Pertinent information from some 3200 fuel cycle RSRs and 1700 non-fuel cycle RSRs has been stored in a computerized data base and used to develop the inventory. Results of the compilation are in disagreement with the two previous estimates for H-3 and for C-14. In particular, non-fuel cycle H-3 inventory for the eight trenches of the present study is approximately a factor of 2 higher than either previous estimate of total site inventory.

Modeling of release processes has been carried out in order to obtain estimates of isotopic release rates from waste packages to the trenches. This modeling is highly speculative, but believed to be state-of-the-art. It required information not only on amounts of the different isotopes, but also on the waste forms and containers holding them. Such information was generally not given on the RSRs and had to be obtained by contact with the generators. Estimated numerical release rate data are given for each trench for H-3, C-14, Cs-137, Sr-90, and Co-60. I-129 is expected to have been totally released within a year of container breaching by corrosion. Most of the Pu, in the form of oxide, will probably not be released at a significant rate. However, about 10%, in the form of soluble nitrate, may be considered to be released on first contact with water.

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*Tables are on microfiche card attached to the inner rear cover of the report.

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EVALUATION OF THE RADIOACTIVE INVENTORY IN, AND ESTIMATION OF ISOTOPIC RELEASE FROM, THE WASTE IN EIGHT TRENCHES AT THE SHEFFIELD LOW-LEVEL WASTE BURIAL SITE

1. INTRODUCTION

1.1 Purpose

In fulfilling its role with respect to regulation of low-level radioactive waste, the Nuclear Regulatory Commission (NRC) requires certain technical information for development of rules, criteria, and other guidance which apply to near-surface disposal of such waste. The Sheffield, Illinois, low-level waste burial site, which was closed in 1978, is considered by NRC to be capable of yielding data of value for NRC's regulatory purposes-of serving, as it were, as a laboratory for obtaining information applicable to other disposal sites, as well as for developing a predictive model of the Sheffield site behavior. Both these functions require detailed knowledge, not only of the amounts and isotopic composition of the buried waste, but also of the waste forms and packaging. These last are essential for estimating release rates of the various isotopes from the waste, which was a specific aim of this study.

The principal source of information on buried low-level nuclear waste is the radioactive shipment record (RSR) which accompanied each shipment made to the burial site. Brookhaven National Laboratory (BNL) has examined the RSRs from 8 of the 17 Sheffield trenches as an aid in developing the detailed inventory necessary to meet the goals of this task. The RSRs occasionally give a reasonably complete picture of the waste, including waste form and package, isotopic composition, and amounts of each isotope. However, the great majority of RSRs give little or no information on waste form and packaging, and many do not give the amounts for individual isotopes. Thus, contact with the shippers of relatively large amounts of waste is required in order to obtain sufficient information for this study.

In the case of the Sheffield burial site, tritium has been found in several places outside the trenches since shortly after the site was closed. In at least one area, it has also been found in small amounts (well below the maximum permissible concentration) at a considerable distance outside the site boundary. It is thus important to try to estimate the amount of H-3 which is likely to be released in the future, and the rate of its release. The same is true for all isotopes of reasonably long half life ($\geq$5 yrs), particularly C-14 and I-129 which, like H-3, are biologically sensitive and are not held up by sorptive processes in the soil. The inventory of H-3 at Sheffield has been considered to be many times greater than that of C-14, but the only quantitative estimates for both isotopes are in wide disagreement.[1,2] Thus, one of the main goals of this study was to prepare an accurate inventory of H-3 and C-14 (as well as other selected isotopes of half life >5 yrs) for eight of

the Sheffield trenches. The inventories are, of course, a minimum requirement for estimating isotopic release rates from the trenches, They also can perhaps provide some insight into methods which might be useful for developing site inventories at other burial sites.

1.2 The Sheffield Site

A description of the Sheffield site as regards historical background, site environmental characteristics, topography, hydrology, and geology can be found in several reports.[1-4] The trench layout is shown in Figure 1.1. The methods used in the construction, filling, and capping of the trenches, together with the repair of caps necessitated by damage from surface water runoff and trench subsidence after closure of the site, are described in Reference 4. Data concerning trench operation (date of operation, volume of waste buried, and amounts of by-product, source, and special nuclear material) reported in Reference 1 are given in Table 1.1. Dimensions of each trench and filling efficiencies are given in Table C-42 of Reference 1. Nuclear Engineering Co., Inc., (NECO), the operator of the site for most of its operating period, reported the volumes and amounts of waste buried each month to the State of Illinois, and this is the source of the data given in the NUS Corp. report,[1] which is reproduced in Table 1.1. In its monthly reports NECO was not required to give a breakdown of the amount of by-product material into amounts of the various isotopes it contained, and, as mentioned previously, one of the principal requirements of this study was to determine the proportions of relevant isotopes in the waste of selected trenches. Thus the only way to obtain this information was to retrieve it directly from the individual shipment records, supplemented if necessary by information from the generators.

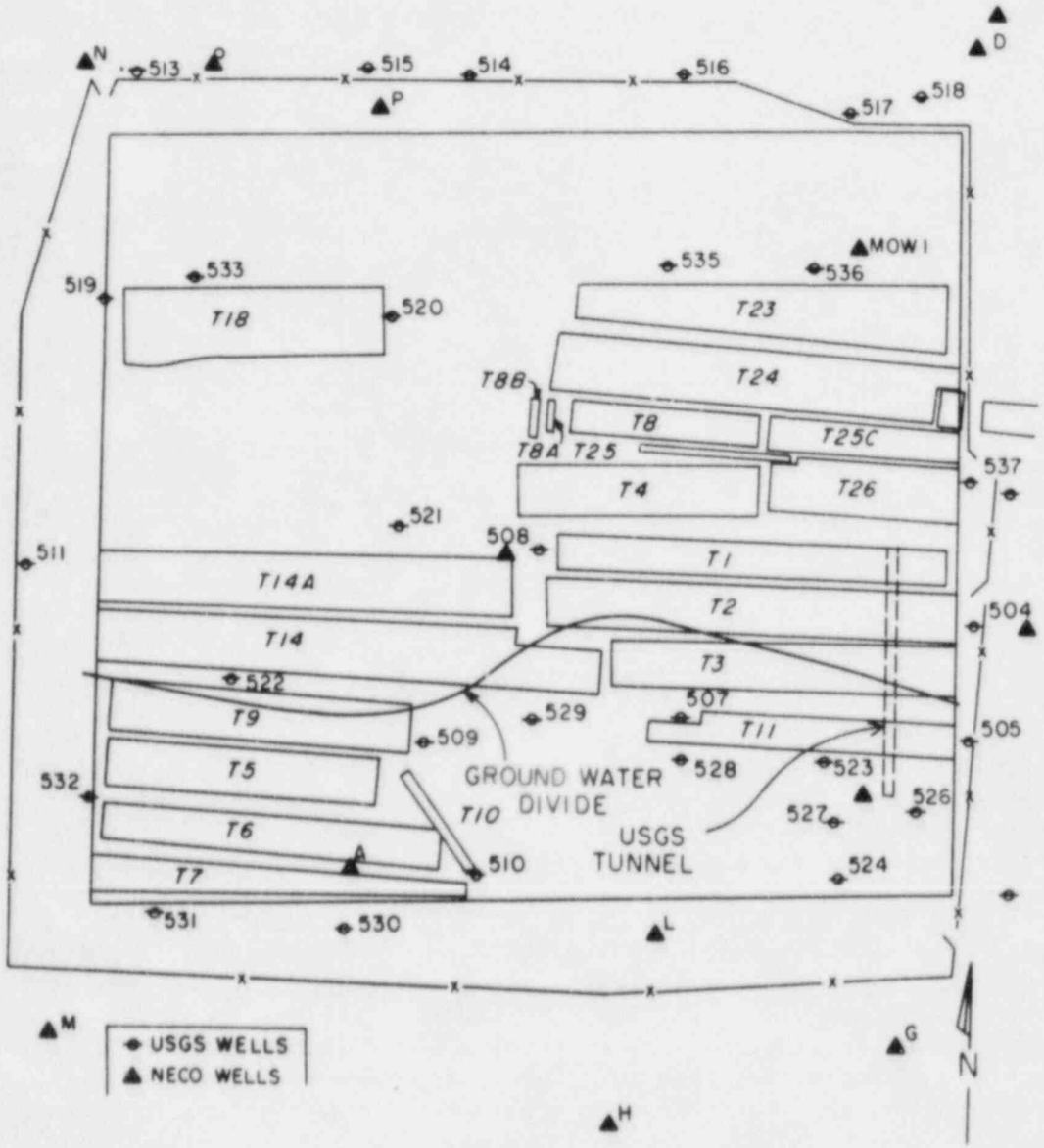

Figure 1.1 Layout of trenches at the Sheffield site. Also shown are the locations of monitoring wells, of the groundwater divide and of the USGS tunnel under Trenches 1, 2, 3, and 11.

Table 1.1

Information on Sheffield Burial Site Trenches[a]

Trench	Period of Operation	Length (ft)	Volume Buried (ft^3)	By-product Material (Ci)	Special Nuclear Material (g)	Source Material (lbs)
1	12/66 - 8/68	450	144,419.16	6,157.30	2,592.28	15,695.46
2	8/68 - 3/71	460	236,23[illegible].14	10,451.15	12,695.86	34,139.79
3	3/71 - 5/72	400	191,200.92	7,758.19	8,339.91	4,546.80
4	5/72 - 4/73	280	197,896.39	4,443.43	4,863.65	3,980.75
5	4/73 - 8/73	350	120,999.13	1,167.66	3,187.33	5,163.95
6	8/73 - 3/74	390	197,365.90	1,327.49	7,040.17	475.32
7	3/74 - 6/74	400	133,709.37	635.76	1,640.73	1,356.00
8	7/74 - 8/74	200	49,364.70	354.96	0.00	0.00
9	7/74 - 2/75	350	185,237.52	1,385.02	912.94	29,613.79
11	12/74 - 6/75	354	93,621.04	1,428.14	683.33	32,897.73
26	5/75 - 8/75	180	166,137.91	991.20	1,087.80	29,611.79
24	6/75 - 5/76	455	305,578.83	5,109.38	4,285.61	24,123.72
25C	4/76 - 8/76	218	65,579.83	863.86	177.58	622.25
18	3/76 -12/76	320	120,655.69	131.30	100.80	198.00
23	8/76 - 1/77	440	180,161.95	4,388.55	211.27	6,577.66
14	1/77 - 9/77	580	394,399.80	7,197.27	1,791.51	293,568.74
14A	8/77 - 4/78	475	351,793.90	6,322.16	4,741.96	118,385.94

[a]Taken from Tables C-41 and C-42 of Reference 1.

1.3 Selection of Trenches for Investigation

The trenches designated for this study were 1, 2, 7, 11, 14A, 23, 24, and 25C (see Figure 1.1 for locations). The reasons for assignment of these particular trenches concern the types and amounts of waste buried in them, their location with respect to known occurrences of radioactivity in soil and water outside the trenches, and their coverage of the geographical area of the site and the time span of site operations. Thus, the selected trenches include those with largely fuel cycle waste and those with largely non-fuel cycle waste. Two of the trenches (7 and 25C) contain rather small amounts of by-product material (Table 1.1), while several contain large amounts (including Trench 2, which contains the largest amount of all the trenches). The operating period of the site is well covered, since Trenches 1 and 2 were operated from the opening of the site for over 4 yrs, Trench 14A was the last Trench to be filled, and the other trenches are spaced throughout the intervening years.

1.4 Organization of the Report

In the following section (Section 2), the methods employed in developing the inventory for each of the designated trenches are described. Detailed inventories for both fuel cycle and non-fuel cycle waste are given for each of the isotopes of concern. Section 3 explains the reasons for contacting shippers regarding waste properties, and discusses those that were chosen for contact. The pertinent information obtained was put in the form of Memos to File, which are included in the report as Appendix B. Based on the amounts of the different isotopes in the waste and the properties of the waste forms and packaging determined from the generator contacts, isotopic release rates to the trenches were estimated. The methods of making these estimates are described in Section 4, and the results are reported by trench and by isotope.

2. INVENTORY OF SELECTED TRENCHES

2.1 Accuracy and Significance of Available Information

2.1.1 Accuracy of Reported Data

It must be stressed at the outset that the inventory developed in this report is based entirely on the records of the waste shippers and the operators of the burial site, and that we have made no judgments regarding the accuracy of the data supplied by the shippers. In general, we have used the information provided on the RSRs exactly as it was given, except for obvious typographical errors, e.g., in naming isotopes. (Figures 2.1 and 2.2, reproduced from the microfilms of the Sheffield RSRs, show an example of one form of RSR used, and the NECO stamp on the reverse used by NECO to summarize the shipment information.) In a few specific instances involving relatively large amounts of radioactivity, the shippers had other documents showing that their RSRs were in error, and the amounts were adjusted accordingly in our data base. An example is the shipment from a university where the RSR listed curie levels of C-14. The university had purchase requistions and invoices showing that only millicurie amounts had been purchased, and the experiments in which the C-14 was used and which gave rise to the waste were known to the Radiation Safety Officer. Another example is that of the generator who shipped large curie amounts of Cs-137 and Sr-90 in lead-lined 30-gal drums. The volumes of the drums were listed for several years as 4.8 ft^3 instead of the correct value of 4.0 ft^3.

There would appear to be no way of knowing how accurate the activity levels listed by the shippers might have been, and we consider it pointless to attempt estimating uncertainties by making assumptions such as, for example, that shippers generally listed larger amounts than they had in order to be conservative. However, it should be emphasized that discrepancies exist between the amounts listed on the RSRs by the shippers and the amounts recorded by the site operators in their summaries of total volumes and radioactivity amounts stamped on the backs of the RSRs.

Unless it was specifically noted that the site operator had contacted the shipper for confirmation or clarification, the amounts of activity listed by the shipper were taken as the proper ones for this study. Occasionally, an arithmetic error was made by burial site personnel in summing the activities of a number of packages. This occurred, for example, with University of Wisconsin RSR 3534, where the total shipment activity given by NECO was 45 mCi, while the actual sum of the packages listed was >28 Ci. Usually the discrepancies consisted of listing millicuries instead of curies, and vice versa. This type of error is understandable since some RSR forms had a column headed "Byproduct Curies," while others were headed "Byproduct Millicuries". In addition, certain shippers who disposed of relatively large amounts of activity, often crossed out the "Milli" in the column heading.

12461

Western Operations Headquarters
P.O. Box 1216
San Ramon, California 94583
(415) 837-1981

P.O. Box 109
Idaho Falls, Idaho 83401
(208) 526-6271

RADIOACTIVE SHIPMENT RECORD

Please complete this form for each shipment and distribute as marked.

CUSTOMER Associated/Searle Corp. DATE Feb. 20, 1976 Page 1 of 1 Pages

ADDRESS 2626 S Clearbrook Dr. Arlington Hgts., Ill. CARRIER

MATERIAL DESCRIPTION 55 gallon drum of solid waste CONTAINER DESCRIPTION

Item no.	Cubic Feet	Gallons Liquid	Physical State	Chemical Form	mRem/hr @ surface	mRem/hr @ 3 ft.	Isotopes	Transport Groups	By-Product (milliCuries)	Source Lbs.	SNM grams	Label Used
1 (HP 29)	7.5	0	Solid		0.3	0.0	^{3}H, ^{35}S, ^{125}I	III, IV	127	0	0	W I
2 (HP 30)	7.5	0	Solid		4.0	1.5	^{123}I, ^{131m}Te, ^{125}I	III, IV	755	0	0	V I
3 (HP 31)	7.5	0	Solid		1.0	0.3	^{3}H, ^{14}C, ^{35}S, ^{125}I	III, IV	345	0	0	V II
4 (HP 32)	7.5	0	Solid		2.0	0.5	^{3}H, ^{32}S, ^{125}I	III, IV	564	0	0	V II
5 (HP 33)	7.5	0	Solid		11.0	3.5	^{3}H, ^{14}C, ^{57}Co, ^{125}I	III, IV	1537	0	0	V III
6 (HP 34)	7.5	0	Solid		2.0	0.4	^{3}H, ^{14}C, ^{125}I	III, IV	138	0	0	V II
7 (HP 36)	7.5	0	Solid		1.0	2.0	^{3}H, ^{131}I	III, IV	245	0	0	V III
8 (HP 21)	7.5	0	Solid		0.2	0.0	^{125}I, ^{35}S, ^{3}H	III, IV	35	0	0	W I
			TOTALS					TOTALS		0	0	

This is to certify that the above named articles are properly classified, described, packaged, marked, and labeled, and are in proper condition for transportation according to the applicable regulations of the Department of Transportation. It is also certified that the articles are in compliance with all regulations applicable at the designated disposal site.

NUCLEAR ENGINEERING COPY

* Reverse side: Terms and Conditions

Feb 19, 1976 (Date) Steven Hontzas (Authorized Signature) Radiation Safety Off. (Title)

Figure 2.1 Enlarged copy of microfilm of one type of RSR used at Sheffield.

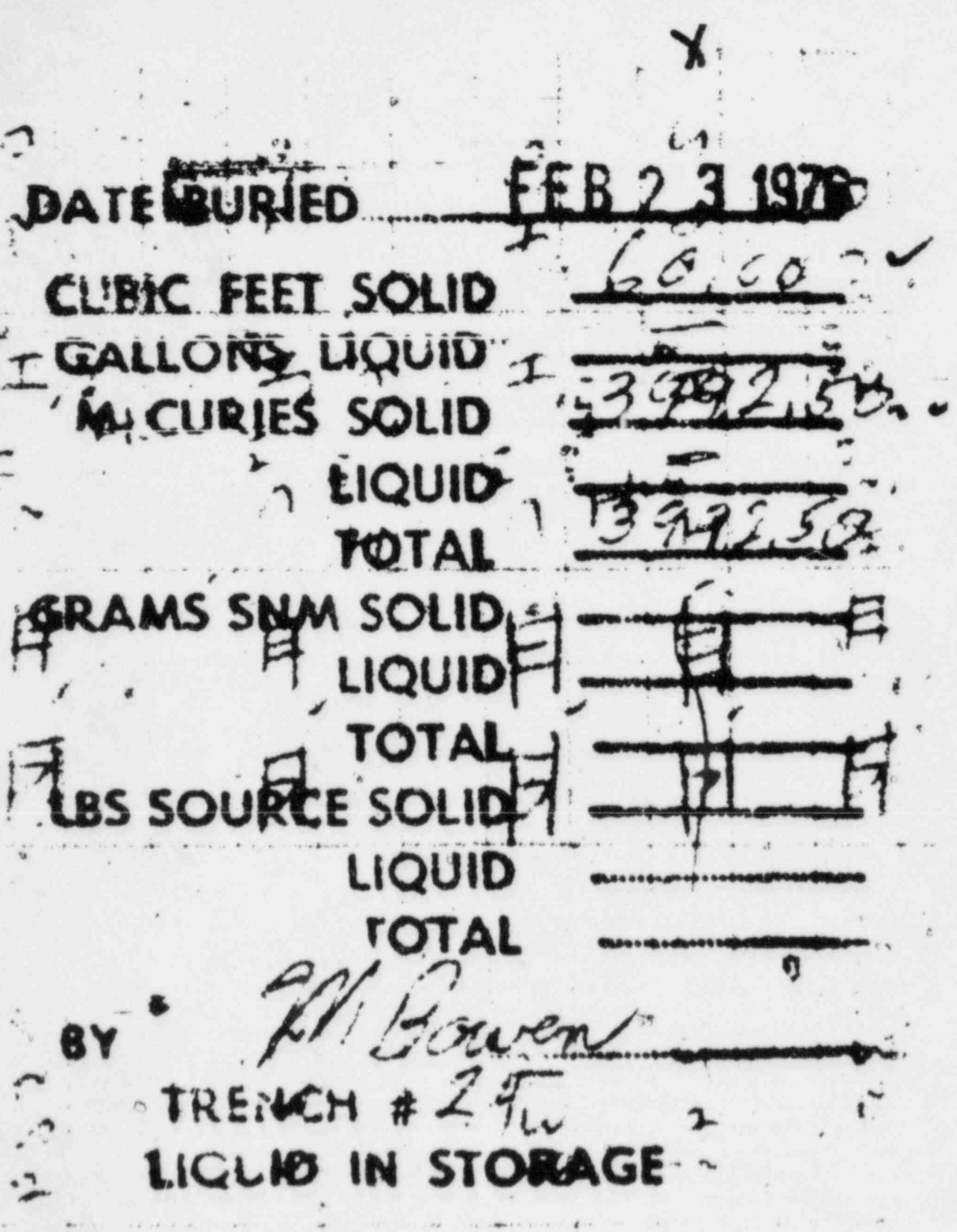

Figure 2.2 Copy of NECO stamp on reverse of RSR shown in Figure 2.1. Overenlarged for clarity.

The largest discrepancy noted was for a shipment of H-3 to Trench 1. The total radioactivity listed by NECO was 11 Ci, whereas, according to the RSR, the shipment actually contained 121 Ci, essentially all of it H-3. Upon contacting the generator, the details given on the RSR were confirmed, so that NECO's inventory would be 110 Ci too low. This is approximately 40% of the trench inventory for H-3, and thus would represent an appreciable error in the source term for Trench 1. Other discrepancies represented only a small percentage of the trench inventory, whether in the case of isotopes found only in non-fuel cycle waste, or in fuel cycle shipments with relatively large amounts of isotopes such as Co-60 and Cs-137. Since the inventories of these latter isotopes were very large, an error of many curies would not result in an appreciable source term error, especially considering the accuracy with which the fuel cycle radioactivities were known.

An uncertainty of a different kind arises whenever a shipper included packages in its shipment which contained a mixture of isotopes and did not give amounts of individual isotopes, but only package totals. This was a frequent occurrence, and the problem it raises is not amenable to a quantitative solution. This is discussed in a later section (Secton 2.3.1.2).

2.1.2 Waste Designated MFP

In addition to the uncertainties in amounts just discussed, there are uncertanties in isotopic composition. As well as the numerous instances where isotopes were listed but no breakdown into amounts of individual isotopes was given, there were many cases of shippers classifying at least a portion of their waste as MFP. While this term means literally mixed fission products, as used by shippers of low-level waste to the Sheffield site, it almost always meant simply a mixture of unspecified radioactive isotopes. A more accurate description of such mixtures used by one shipper was UBGER, standing for unspecified beta gamma emitting radioisotopes. Even when used by utilities shipping fuel cycle waste, the designation MFP had no precise meaning. Although their waste contained fission products, it also had a high proportion of activation products.

As used by non-fuel cycle shippers, the term is even less meaningful, since fission products may or may not have been present in their waste. In general, no specific isotopes can be listed for such waste, and we have had to omit it from consideration in development of release scenarios. In the case of Argonne National Laboratory (ANL), the shipper of by far the largest amount of waste designated MFP, this is true for most of the trenches. However, for Trenches 1 and 2, a large proportion of the waste was contained in two series of special shipments of rather high radioactivity content (108 Ci in that portion sent to Trench 1 and 1346 Ci in the portion sent to Trench 2). This waste did contain fission products and activation products, and a rough estimate can be made of isotopic composition for that part of ANL's contribution (see Section 2.3.2.4).

2.2 Compiling the Data

2.2.1 Use of Data Base

The requirements of this study for detailed information on many different aspects of the waste and the burial trenches suggested the use of a computerized data base as the most efficient way to utilize the contents of the Sheffield RSRs. Such a data base would contain the data in an easily accessible and retrievable form, and it could also be structured so that interrelationships between various elements of the data could be easily established. Use of the data base management system also makes it possible to generate lists, counts, and sums of the data and to present the data in tabular form. These features greatly facilitate analysis and reporting of the results.

2.2.2 Description of the Data Base

The data base chosen was set up on BNL's CDC 6600 computers using Intel Corporation's System 2000 general-purpose data base management system. A complete description of this system is given in a manual published by the Intel Corporation.[5] For the present, it suffices to know that a data base defined under System 2000 establishes components for each data element, arranges the components into groups (records) of related elements and places these groups of data into a hierarchical structure. A diagram of the structure of the current data base is shown in Figure 2.3.

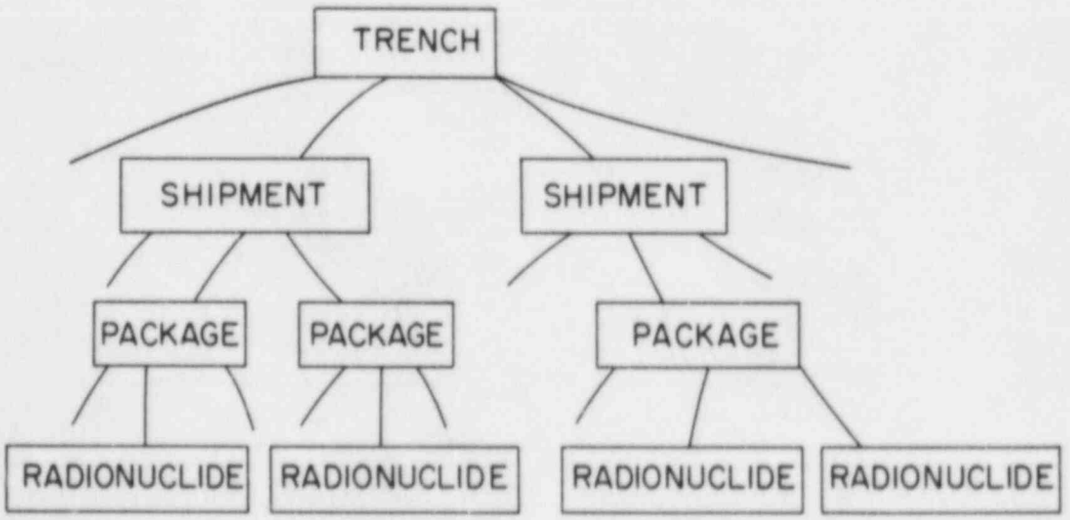

Figure 2.3 Structure of the data base.

From the figure, it is seen that the records in this data base are arranged into four levels. The first level contains information about the trenches, the second level contains information about the various shipments to these trenches, the third level contains information about the packages in these shipments, and the fourth level contains information about the radionuclides in these packages. Also, a description of the data base schema is given in Figure 2.4. This schema is the collection of component definitions for each of the data elements in the data base. In the schema, the various levels of the data are identified by increasing indentations.

```
DESCRIBE%

 SYSTEM RELEASE NUMBER        M2.80D
 DATA BASE NAME IS SHEFFIELD
 DEFINITION NUMBER       8
 DATA BASE CYCLE       834
     1*  TRENCH (CHAR XXXX)%
     2*  OPENDT (DATE)%
     3*  CLOSEDT (DATE)%
    10*  SHIPMENT (RECORD)%
      11*  SHIPMT DT (DATE IN 10)%
      12*  ORIGIN (CHAR X(25) IN 10)%
      13*  ADDRESS (NON-KEY TEXT X(7) IN 10)%
      14*  SHPMT NUM (CHAR X(10) IN 10)%
      15*  SHPMT DESGN (CHAR X(10) IN 10)%
      18*  SOLID VOL (DECIMAL NUMBER 9(6).9 IN 10)%
      19*  LIQUID VOL (DECIMAL NUMBER 9(6).9 IN 10)%
      20*  RADIOAC (DECIMAL NUMBER 9(6).999 IN 10)%
      21*  WTSOLSNM (DECIMAL NUMBER 9(6).99 IN 10)%
      22*  WTLIQSNM (DECIMAL NUMBER 9(6).99 IN 10)%
      23*  WTSOLSRC (DECIMAL NUMBER 9(6).99 IN 10)%
      24*  WTLIQSRC (DECIMAL NUMBER 9(6).99 IN 10)%
      25*  COMMENT (TEXT X(40) IN 10)%
      29*  ACTIVITY RATIO (DECIMAL NUMBER 9(6).999 IN 10)%
      30*  PACKAGE (RECORD IN 10)%
        31*  PKGNO (INTEGER NUMBER 999 IN 30)%
        32*  WTSOURCE (DECIMAL NUMBER 9999.99 IN 30)%
        33*  WTSNM (DECIMAL NUMBER 9999.99 IN 30)%
        34*  WSTFORM (CHAR X(10) IN 30)%
        35*  CHEMFORM (CHAR X(16) IN 30)%
        36*  TOTRADIA (DECIMAL NUMBER 9(6).999 IN 30)%
        40*  RADIONUC (RECORD IN 30)%
          41*  NUCLIDE (CHAR X(30) IN 40)%
          42*  AMOUNT (DECIMAL NUMBER 9(6).999 IN 40)%
---
```

Figure 2.4 Computer printout of the schema for Data Base Sheffield. The legend is given on page 11 for the sake of clarity.

Legend for Figure 2.4

OPENDT	date trench was opened
CLOSEDT	date trench was closed
SHIPMT DT	date of shipment
SHPMT NUM	RSR number
SHPMT DESGN	shipment designation according to waste category
SOLID VOL	volume of solid waste in shipment, in cubic feet
LIQUID VOL	volume of liquid waste in shipment, in gal
RADIOAC	total shipment radioactivity, in mCi
WTSOLSNM	total weight of solid special nuclear material (SNM), in g
WTLIQSNM	total weight of liquid SNM, in g
WTSOLSRC	total weight of solid source material, in lb
WTLIQSRC	total weight of liquid source material, in lb
ACTIVITY RATIO	20* divided by 18*, in mCi/cubic foot
PKGNO	package number
WTSOURCE	weight of source material in package, in lb
WTSNM	weight of SNM in package, in g
WSTFORM	physical state (solid, liquid, or gas) of the waste in the package
CHEMFORM	chemical form of the waste in the package
TOTRADIA	total radioactivity in the package, in mCi
RADIONUC	radionuclides in the package
AMOUNT	amount of radioactivity, in mCi, for the nuclide listed in 41*

2.2.3 Inputting the Data

2.2.3.1 Selection of the Data

Two principal criteria were used to select the RSRs from which data were to be input. The first was that the shipment had to contain isotopes of half-life $\geq$5 yrs in order to be selected. Specifically, the isotopes of concern, or relevant isotopes, are considered to be those listed in Table 3.8 of Volume 2 of the report, "Data Base for Radioactive Waste Management."(6) The second criterion set minimum values for the amounts of radioactivity which qualified a shipment for inclusion.

All shipments containing no relevant isotopes were omitted. All shipments containing isotopes of concern and which satisfied the second criterion regarding amount of radioactivity were processed, but in general only those packages within a given shipment that contained relevant isotopes were listed in the package record (numbers 31 to 42 in the schema). That is, packages containing only isotopes of no concern were omitted.

For the first five trenches processed (7, 11, 14A, 23, and 25C), the minimum amount of raidoactivity set by the second criterion was 1 mCi for non-fuel cycle waste and 10 mCi for fuel cycle waste. Analysis of the data obtained from these trenches showed that very small shipments contributed an insignificant proportion of the total waste in a trench, so that the minimum was raised to 10 mCi for non-fuel cycle waste and 100 mCi for fuel cycle waste during inputting of the Trench 1 data. These limits thus apply to part of Trench 1 and all of Trenches 2 and 24. To avoid putting unnecessary information in the data base, a limitation was also placed on the inclusion of package information. Individual packages in selected shipments were omitted if the activity of relevant isotopes was only a small fraction of a millicurie.

The only exceptions to the second criterion were made for I-129. This biologically sensitive isotope, like H-3 and C-14, is not held up by the soil, but moves with the groundwater, so it is of concern to know how much I-129 was buried in the trenches and at what rate it is likely to be released to the trenches from its containers. All packages containing I-129 were therefore included in the data base, regardless of the curie amount of the package or of the shipment in which it was contained.

2.2.3.2 Source Material and Special Nuclear Material

Information for source material and special nuclear material (SNM) was entered into the data base for the first five trenches studied (7, 11, 14A, 23, and 25C). During inputting of Trench 1 data, it was decided not to continue entering information regarding source material and SNM shipments, unless the SNM consisted of, or contained, plutonium. Release rates of thorium and uranium were not required in this study, thus no information on their shipment was put in the data base for Trenches 2 and 24. However, when the RSRs for these trenches were being scanned, the shipments of source material and SNM were listed along with the by-product shipments, so that if it is ever decided to put their information into the data base, the RSR numbers will be readily available. They are listed, by trench, in Appendix A.

2.2.3.3 Procedure for Inputting

Copies of the RSRs on microfilm were made available by US Ecology, formerly Nuclear Engineering Co. (NECO), operator of the Sheffield site during most of its operating life. The rolls of microfilm were initially scanned by members of the scientific staff to select the RSRs to be entered into the data base, using the criteria discussed in the previous Section (2.2.3.1). The scientific staff also designated each selected RSR as fuel cycle or non-fuel cycle. In a few cases where it was not certain to which category it belonged, the shipment was designated as "other". Lists of the selected RSRs were then compiled, and both scientific and clerical staff entered the pertinent information into the data base directly from the microfilmed records. After each day's entries were checked by the scientific staff, a permanent record of the updated data base was made on magnetic tape.

During scanning of the microfilm and compilation of the lists of selected RSRs, all non-fuel cycle shipments containing amounts of relevant isotopes ≥1 Ci were noted and made into a list for each trench. After the selected data for a given trench had been entered into the data base, the non-fuel cycle shipments of ≥1 Ci were printed out and compared with at least the ≥1 Ci list obtained during scanning, and often also with the microfilm record. Any errors found in the data base were then corrected. This procedure ensured that all the non-fuel cycle shipments containing ≥1 Ci of relevant isotopes were entered correctly in the data base. These shipments were found to contribute of the order of 90%, and usually more, to the total non-fuel cycle inventory of relevant isotopes. In addition to this check, many random checks between the data base and the microfilm were made in the course of checking other computer-generated lists, such as lists of shipments containing specific isotopes. Because a large proportion of fuel cycle shipments were >1 Ci, and a very high fraction were listed merely as containing MFP or Co-60 and Cs-137, fuel cycle shipments were not checked in the same way as non-fuel cycle. Deliberate random checks were made, and several series of unusually large shipments which were noted from time to time were checked against the microfilmed records.

2.3 Inventory Compilation

The inventory results obtained from the data base are presented in the following sections under two main headings, fuel cycle and non-fuel cycle. The information is further broken down into data on individual isotopes, and also by trench, since each trench has to be considered separately in Section 3 of the report, which deals with the release of individual isotopes to the trench. Before presenting the results, some problems involved in the development of the inventory are discussed.

2.3.1 Potential Errors in Inventory Data

2.3.1.1 Pitfalls Encountered in Use of the Data Base

The way in which package information is entered into the data base is very important in enabling it to be retrieved readily and utilized when searches are conducted for making lists and doing counts and summations. For example, information pertaining to the isotopic composition of the waste may for some reason be put into the comments (C25 in the schema of Figure 2.2), and, to avoid duplication, the isotopes may not be listed in the package information under C41 of the schema. Since lists, counts, and sums involving a particular isotope are generated using C41 entries for that isotope for the computer search, shipments for which no C41 entries are made will be missed in such a search.

This form of inputting occurred a number of times while the non-fuel cycle data were being entered, and was first discovered to be a problem when a large shipment was missed when checking the computer record of shipments >1 Ci against that obtained during manual scanning of the microfilmed RSRs. The problem is manageable, since the required information is in the comments,

but part of the advantage of having a data base is lost. After the non-fuel cycle data for the first five trenches (7, 11, 14A, 23, and 25C) had been input, it was determined that a C41 entry always be included in the package information. Thus, there should be no problem from this source for work with the last three trenches (1, 2, and 24) and for the fuel cycle data.

To determine what proportion of the waste was affected, lists were made of the information on shipments in which H-3 was mentioned in C25, for Trenches 7, 11, 14A, 23, and 25C. These lists were compared with printouts of the shipments where H-3 was entered in C41 to obtain the shipments where it was mentioned only in C25. As an example of the information in the data base for these latter shipments, the list for Trench 11 is given in Table 2.1. The Trench 11 list had no unusual features and was chosen merely because it contained a reasonable number of shipments. The amounts of waste containing H-3 were summed for the two categories of shipment - the category of all shipments containing $\leq$1 Ci, and that of the shipments for which H-3 was mentioned only in C25.

Table 2.1

Shipments to Trench 11 in which H-3 Mentioned Only in Comments

Shipper	RSR	Date	Amount Alone (mCi)	Amount Mixed (mCi)
Mobay Chemical	----	2/06/75		655
Harper Hospital	1628	2/10/75		6
Kallestad Labs	2058	2/26/75		11.8
Harper Hospital	1492	3/10/75		6
VA Hospital	6082	3/11/75	32	
Abbott Labs	1616	3/13/75		8.5
Kallestad Labs	2060	3/25/75		9.3
Radiation Monitoring Service	0898	3/27/75	8	
Harper Hospital	01491	4/07/75		5
Marshfield Medical Foundation	11693	4/10/75		1.5
Kallestad Labs	2062	4/22/75		14.7
Harper Hospital	1490	5/05/75		5
Southern Illinois	11066	5/13/75		16
Kallestad Labs	2064	5/20/75		273

The results are given, by trench, in Table 2.2. Roughly 1/6 of the shipments were affected, and roughly the same fraction of activity. Trench 14A had the largest number of shipments mentioned only in C25, and also the

largest amount of H-3 affected, approximately 6 Ci. To put this in perspective, the total H-3 inventory for the trench was over 700 Ci, so the fraction affected is less than 1%. Although the inventories of the other trenches are much smaller, so are the amounts affected, so the percentages remain small and will cause insignificant errors in the overall trench inventories. The same situation exists for C-14, and may to a small extent exist for other isotopes.

Table 2.2

Information on Shipments in Which H-3 Mentioned Only in Comments

	Shipments With H-3 Only in C25			All Shipments <1 Ci Containing H-3	
Trench	Number of Shipments	H-3 Alone (Ci)	Mixed With Other Isotopes (Ci)	Number of Shipments	Amount of Waste (Ci)
7	18	0.10	0.86	61	7.4
11	14	0.04	1.02	83	7.38
14A	39	0.87	5.04	165	17.78
23	29	0.01	3.11	101	11.88
25C	2	0	0.49	54	5.85
Totals	102	1.02	10.52	464	50.29

Another example of difficulty in retrieving information occurs when not all values for package activity, C36, are entered for a shipment. In cases where a number of packages were identical, or very similar (e.g., same isotope or isotopes), only one package would be given in detail, as an example, and this fact would be noted in the comments (C25). Thus again, as with the shipments where C41 information was not entered, reference to the comments is required to obtain the full picture. Also the situation only arises for shipments $\leq$1 Ci since shipments >1 Ci have all been cross-checked. In most cases, the amounts entered in C20 represent the sums of the C36s for the packages in question, but, if not, reference will have to be made to the comments in C25 to obtain the correct sums. There is no problem with fuel cycle waste since, for most fuel cycle shipments, all packages in the shipment had the same C41s (very often simply MFP), and C36 values were of little or no interest - in fact, the sum of C36s for such shipments was essentially always equal to C20. In the few instances where a shipment contained different kinds of packages, package details were given, including the C36 values.

In one instance in the entire data base, a number was too large to be entered. When the data base was set up, it was not anticipated that there would be any single shipments with a total radioactivity greater than 1000 Ci, so a maximum of six digits before the decimal place was permitted in C20. A

shipment to Trench 2 (RSR 3738, March 18, 1971) contained 1455 Ci (1,455,000 mCi), so this value for its total activity could not be entered. The value used was the actual value divided by 10, and a notation was made in the comments (C25) to multiply the value given in C20 by 10 to obtain the correct value for the total radioactivity of the shipment.

2.3.1.2 Uncertainties for Packages of Isotope Mixtures

In general, whenever a non-fuel cycle shipper put mixtures of isotopes into its packages and did not provide a breakdown of the amounts of each isotope, there is now no way of determining what these amounts were. Thus, some estimate has to be made of the isotopic distribution in such cases. This is a different situation than that discussed in the section on waste designated MFP (Section 2.1.2). For the usual use of the MFP designation with non-fuel cycle waste, no estimate of isotope amounts is possible. However, when isotopes are at least identified, estimates of their amounts can often be made. The situation with regard to fuel cycle shipments is a special case, and will be discussed in the section dealing with fuel cycle waste (Section 2.3.3).

The accuracy of estimates in the case of non-fuel cycle shipments is crucial only for large shipments (of the order of a few Ci and larger) where the information is important for use in release scenarios. There turned out to be few (<50) large packages of isotope mixtures with no breakdown by isotope, since a generator with a large amount of a particular isotope to dispose of, usually shipped it in a container by itself. The few large packages of mixed isotopes almost always involved H-3, with C-14 or I-125 or I-131 as the other isotope. Only 3 or 4 instances involving other isotopes of concern were observed.

When shippers of large curie amounts were contacted for further information on waste form and isotopic distribution, they were often able to shed some light on the isotopic distribution of their more usual low-activity waste. While not crucial to the overall H-3 inventory, since shipments <1 Ci contributed <10% for most trenches, the information on low-level shipments allowed useful estimates to be made for that component of the inventory.

From discussions with shippers, a reasonably consistent picture emerged for shipments of packages containing mixtures of H-3 and C-14 in which the amounts of each isotope were not identified. Copies of Memos to File concerning these discussion are included, with others about waste form and packaging, in Appendix B. Of those shippers asked specifically about isotopic distribution, all stated that their mixtures contained mostly H-3. One shipper gave a rough estimate of 80% H-3, while another estimated well over 90%, and another said that its mixture was nearly all H-3. Our estimates have been based on the values given us by the shippers.

For more complex mixtures, the picture is not clear, since there were a number of isotopes involved whose amounts were not well known. Some of these isotopes might generally be present in greater amounts than H-3, while others would be in smaller amounts. Almost all of the shipments in this category involved relatively small total activities, so uncertainties in H-3 and C-14 inventories would not be appreciable regardless of the assumption made about amounts of individual isotopes. We have assumed equal distributions in the case of these complex mixtures, where most of the isotopes other than H-3 and C-14 are isotopes of no concern such as I-125, I-131, P-32, S-35, Cr-51, Zn-65, Se-75, etc.

2.3.2 Non-Fuel Cycle Inventory

2.3.2.1 Tritium

Of the isotopes of interest to this study, tritium ranks next to Co-60 and Cs-137 in terms of amount buried in the whole Sheffield site. Also, as previously pointed out, it is of special interest since it is biologically sensitive, and is not held up by interaction with the soil. Because it is not held up, it has been found in groundwater samples taken from wells outside the trenches.[2] On the following pages, we present compilations of the shipments $\geq$1 Ci to each trench, together with other information concerning small shipments and total trench inventories. In Tables 2.3 to 2.10 of shipments $\geq$1 Ci, information on waste form is included where it was given on the RSR. Usually nothing more was given than the physical state of the waste. Table 2.11 gives the estimated total H-3 inventory by trench.

In Table 2.12 , the H-3 trench inventories are compared with total trench inventories (taken from Reference 1) for the eight trenches surveyed in this study. The number of shipments containing H-3 for each trench is also given. The amounts of H-3 in the trenches follow the total amounts of by-product material in a rough way. The reason for this rough correspondence presumably is because of the large number of shipments containing H-3. All the other non-fuel cycle isotopes of concern except C-14 were contained in relatively few total shipments, including shipments of $\geq$1 Ci. Thus, on a statistical basis, large fractions of the total inventory could turn up in any trench.

Table 2.3

Non-Fuel Cycle H-3 Shipments >1 Ci to Trench 1

Shipper	RSR	Date	Amount (Ci) and Form
Indiana University	203	2/28/67	1.0, solid
Abbott Laboraties	743,4	3/10/67	13.7, gas
University of Wisconsin	----[a]	6/15/67	4.6, solid
Abbott Laboratories	----	6/16/67	4.5, gas
G. D. Searle	----	6/30/67	1.7, solid
Luminous Processes, Inc.	----	7/11/67	135.9, solid, Luminous scrap dials and wipe tissues
University of Wisconsin	----	9/07/67	3.8, solid
3M Co.	----	12/20/67	1.02, solid
US Public Health Service	----	1/22/68	13.0, solid
University of Wisconsin	----	3/08/68	121, solid
US Public Health Service	----	6/04/68	2.6, solid
Chicago Medical School	----	6/14/68	2.0, solid, C-14 and H-3 mixed
University of Minnesota	----	7/09/68	1.0, solid

[a]No entry under the RSR heading means that the form used for that shipment had no number, a common occurrence in the first year or two of Sheffield's operation.

Table 2.4

Non-Fuel Cycle H-3 Shipments >1 Ci to Trench 2

Shipper	RSR	Date	Amount (Ci) and Form
University of Wisconsin	----	9/04/68	5.0, solid
Mallinckrodt	----	9/10/68	<7, solid
University of Illinois	----	11/06/68	9.5, solid
Abbott Laboratories	----	12/26/68	3.0, solid
ITT Industrial Labs	----	12/30/68	2.0, solid, ampule sealed on vacuum system manifold
University of Missouri	----	1/13/69	1.7, solid
Amersham/Searle	----	2/04/69	5.0, solid
University of Wisconsin	11206	2/04/69	1.7, solid
Unknown	----	2/10/69	2.6, solid
Purdue University	----	2/26/69	2.3, solid
Purdue University	11245	3/13/69	6.0, solid
University of Wisconsin	11212	3/20/69	83.0, solid
Purdue University	11247	3/27/69	<6.0, solid
University of Missouri	----	4/07/69	1.0, solid
Abbott Laboratories	----	4/17/69	1.7, solid
Purdue University	11249	4/24/69	5.0, solid
Purdue University	11250	5/15/69	5.0, solid
University of Wisconsin	11211	5/22/69	124.5, solid
Nuclear Chicago	----	5/27/69	6.6, solid
University of Illinois	----	6/13/69	1.6, solid
Purdue University	11252	6/26/69	5.0, solid
US Radium Corp.	11543	6/27/69	<82, solid, one package, also contains Kr-85, amounts not specified
University of Missouri	----	7/28/69	1.65, solid
University of Wisconsin	11207	7/22/69	1.7, mostly solid
Caterpillar Tractor Co.	11652	9/18/69	<4.8, tritiated oil absorbed on vermiculite
Atomic Disposal Co.	----	9/22/69	<8.2, solid
University of Wisconsin	11208	9/25/69	6.6, mostly solid
Amersham/Searle	4537	10/22/69	4.0, solid
Purdue University	11257	10/30/69	5.0, solid
Illinois Institute of Technology	4132	11/14/69	7.0, solid
Purdue University	11259	12/16/69	1.8, solid
Parke Davis and Co.	3457	12/18/69	2.4, solid

Table 2.4, Continued

Non-Fuel Cycle H-3 Shipments >1 Ci to Trench 2

Shipper	RSR	Date	Amount (Ci) and Form
Caterpillar Tractor Co.	5521	12/22/69	≤5.0, presumably liquid absorbed on vermiculite[a]
University of Wisconsin	3532	1/29/70	16.1, solid
Mead Johnson Research Center	3674	2/06/70	2.5, liquid
Parke Davis and Co.	3458	2/25/70	1.1, solid
Amersham/Searle	3763	3/06/70	1.8, solid
Miles Laboratories	----	3/19/70	<2.0, solid
University of Missouri	39834	3/25/70	4.8, solid
Atomic Disposal Co.	3504,5	3/30/70	<3.6, solid
Caterpillar Tractor Co.	11116	4/02/70	≤5.8, presumably liquid absorbed on vermiculite[a]
Purdue University	3671	4/07/70	5.0, solid
Purdue University	3673	4/19/70	30, solid
University of Wisconsin	----	4/29/70	6.6, solid
Michigan State University	3548	6/25/70	25, gas
Amersham/Searle	3764	7/17/70	2.5, solid
Caterpillar Tractor Co.	5522,3	8/18/70	≤5.8, presumably liquid absorbed on vermiculite[a]
Purdue University	3652	8/28/70	25.0, solid
University of Missouri	3985,6	9/25/70	1.8, solid
University of Wisconsin	----	10/08/70	17.1, solid
Amersham/Searle	3765	10/16/70	3.0, solid
University of Illinois	4257	11/03/70	1.2, solid
Purdue University	3656	12/01/70	17.1, solid
Washington University	4075	12/22/70	19.6, solid
University of Missouri	11646	1/26/71	3.3, solid
Mead Johnson Co.	10047	1/28/71	7.0, solid
Luminous Processes Inc.	9654	2/19/71	29.0, solid
University of Wisconsin	3534	2/23/71	28.0, solid
Atomic Disposal Co.	4083	3/16/71	31.5, 29 Ci solid 2.5 Ci liquid

[a]Listed as liquid on RSR, but as solid by NECO, so presumably similar to Caterpillar Tractor's RSR 11652 of 9/18/69.

Table 2.5

Non-Fuel Cycle H-3 Shipments >1 Ci to Trench 7

Shipper	RSR	Date	Amount (Ci) and Form
Mallinckrodt Nuclear	----	3/04/74	9.6, liquid
Ames Laboratory	10375	4/10/74	13, solid
Illinois Institute of Technology	2565	4/19/74	6.5, solid
Amersham/Searle Corp.	11913	5/10/74	<2.0, solid, mixed with other isotopes

Table 2.6

Non-Fuel Cycle H-3 Shipments >1 Ci to Trench 11

Shipper	RSR	Date	Amount (Ci) and Form
University of Illinois	1726	4/02/75	1.05, solid
Michigan State University	2528	4/09/75	<2.2, solid, mixed with C-14
Shell Development Corp.	10571	4/15/75	1.2, solid
G. D. Searle and Co.	2634	4/18/75	4.4, solid
3M Co.	1792	4/23/75	<8.3, solid, mixed with C-14
WARF Institute	10574	4/23/75	<1.1, liquid, mixed with C-14
Ames Laboratory	03314	4/30/75	7, solid
Dow Chemical Co.	2282	5/01/75	8, solid

Table 2.7

Non-Fuel Cycle H-3 Shipments >1 Ci to Trench 14A

Shipper	RSR	Date	Amount (Ci) and Form
Brookhaven National Laboratory	9421	8/09/77	1.5, solid
Dow Chemical Company	22527, etc.	8/16/77	6.7, solid
Brookhaven National Laboratory	---	8/22/77	1.8, solid
Brookhaven National Laboratory	---	8/25/77	40.3, solid
Brookhaven National Laboratory	8311,2	9/07/77	144.8, solid
Brookhaven National Laboratory	8313,4	9/13/77	95.8, solid
Brookhaven National Laboratory	8315,6	9/13/77	1.4, solid
Pathfinder Laboratory,	29541	9/13/77	1.0, solid
Luminous Processes, Inc.	35405	9/16/77	29.9, solid, "tritiated zinc sulfate"[a]
Brookhaven National Laboratory	8309,10	9/21/77	21.5, solid
Brookhaven National Laboratory	8324	9/22/77	128.9, solid
Atomic Disposal Co.	36854,-6	9/30/77	<1.5, mostly solid, animal carcasses
Endocrine Laboratories of Madison, Inc.	19705	10/11/77	1.9, solid, most (1.8 Ci) in cement
University of Wisconsin	36729,30	10/13/77	<21.2, mostly solid
Luminous Processes, Inc.	35406	10/28/77	29.9, solid, "tritiated zinc sulfate"[a]
Luminous Processes, Inc.	35407	11/17/77	29.9, solid, "tritiated zinc sulfate"[a]
Amersham Corp.	23334	12/12/77	<9.1, solid, water-based radiochemicals
Luminous Processes, Inc.	35408	12/22/77	29.9, solid, "tritiated zinc sulfate"[a]
Westinghouse Electric Corp.	08689	1/06/78	4.8, solid, Ti targets
Dow Chemical Co.	19978	1/19/78	1.02, solid
Radiation Control and Radiological Services	---	1/27/78	1.5, solid, mixed organics
Luminous Processes, Inc.	35409	1/30/78	29.9, solid, "tritiated zinc sulfate"[a]
Amersham Corp.	23335	2/03/78	<1.6, solid, water-based radiochemicals
University of Illinois	50986	2/14/78	5.0, solid, NOS lab waste
Illinois Institute of Technology	---	2/15/78	74.5, solid
Midwest Research Institute	32483	3/14/78	2.3, solid, animal carcasses
Amersham Corp.	51143	3/31/78	<2.6, solid, water-based radiochemicals

[a]As listed by shipper; presumably should be sulfide rather than sulfate.

Table 2.8

Non-Fuel Cycle H-3 Shipments >1 Ci to Trench 23

Shipper	RSR	Date	Amount (Ci) and Form
Midwest Research Institute	32482	8/09/76	4.0, solid
Luminous Processes, Inc.	11108	8/24/76	37.4, "tritiated zinc sulfate"[a]
Luminous Processes, Inc.	11107	8/27/76	29.9, "tritiated zinc sulfate"[a]
Amersham/Searle Corp.	31689	9/09/76	<3.5, solid, water-based radiochemicals
Purdue University	33357-8	9/29/76	2.7, solid
Timex Corp.	33457	10/01/76	25, "poly/styrene"[b]
3M Company	20075, etc.	10/05/76	<5.5, mostly solid, radio-labelled organics
Luminous Processes, Inc.	11109	10/15/76	29.9, "tritiated zinc sulfate"[a]
USAF, Kelly AFB	31982	10/21/76	<13.6, solid
Amersham/Searle Corp.	31692	10/22/76	<3.3, solid, water-based radiochemicals
Ames Laboratory	22159-60	10/26/76	2.5, solid
University of Wisconsin	12447	10/28/76	<5.3, (<3.1 Ci solid, <2.2 Ci liquid)
Mobay Chemical Corp.	12572	12/02/76	<1.7, solid
Luminous Processes, Inc.	11091	12/03/76	30.0, "tritiated zinc sulfate"[a]
Abbott Laboratories	29613	12/03/76	1.0, solid
Amersham/Searle Corp.	31693	12/03/76	<1.9, solid, water-based radiochemicals, lab waste

[a]As listed by shipper; presumably should be sulfide rather than sulfate.
[b]As designated by shipper.

Table 2.9

Non-Fuel Cycle H-3 Shipments ≥ 1 Ci to Trench 24

Shipper	RSR	Date	Amount (Ci) and Form
University of Missouri	--	9/04/75	1.0, solid
3M Co.	20031	9/10/75	1.1, solid
University of Notre Dame	10591	10/20/75	1.0, solid
University of Wisconsin	12439,40	10/23/75	<7.4, 5 Ci solid, <2.4 Ci liquid
GM Corp.	21375	10/28/75	2.9, solid
Luminous Processes, Inc.	11099	11/07/75	36, solid, tritiated ZnS[a]
Amersham/Searle	12460	11/13/75	<2.6, solid
University of Illinois	20262,3	12/08/75	7.7, solid
Upjohn Co.	01222	12/17/75	6.5, solid
Dow Chemical Co.	19695	12/18/75	1.6, solid
Luminous Processes, Inc.	11100	12/22/75	45, solid, tritiated ZnS[a]
Upjohn Co.	01224	2/11/76	6.9, solid
Luminous Processes, Inc.	11101	2/13/76	40.4, solid, tritiated ZnS[a]
Atomic Disposal Co. (ADCO)	20312,etc.	2/16/76	<5.1, solid
Amersham/Searle	12461	2/20/76	<3.2, solid
Luminous Processes, Inc.	11102	3/19/76	49.4, solid, tritiated ZnS[a]
Honeywell SAPD	21030	3/23/76	1.5, solid, paint
Luminous Processes, Inc.	11103	4/09/76	36, solid, tritiated ZnS[a]
KMS Fusion	23323	5/5/76	≤5, solid
Luminous Processes, Inc.	11104	5/13/76	49.4, solid, tritiated ZnS[a]

[a] Listed as "Tritiated Zinc Sul."

Table 2.10

Non-Fuel Cycle H-3 Shipments >1 Ci to Trench 25C

Shipper	RSR	Date	Amount (Ci) and Form
University of Wisconsin	12444,5	5/27/76	<12.9, (1.1 liquid, <11.8 solid)
Amersham/Searle Corp.	12462	5/28/76	<5.5, solid, water-based radiochemicals
Luminous Processes, Inc.	11105	6/11/76	36.0, solid, "tritiated zinc sulfate"[a]
University of Missouri	---	6/15/76	2.7, mostly solid
Amersham/Searle Corp.	12463	6/16/76	<2.1, solid, water-based radiochemicals
Midwest Research Institute	21201	6/17/76	7, contaminated lab-ware in drums
University of Missouri	21758	6/23/76	8.0, "T_3O"[b] in Toluol
Luminous Processes, Inc.	11106	7/02/76	31.5, solid, "tritiated zinc sulfate"[a]
University of Illinois	20278,9	7/07/76	1.7, liquid (toluene and xylene)
Marshfield Medical Foundation	21178,9	7/21/76	3.5, liquid (alcohol)
Abbott Laboratories	23650	7/23/76	<2.8, solid
Miles Laboratories, Inc.	22633	7/26/76	1.8, liquid

[a]As listed by shipper; presumably should be sulfide rather than sulfate.
[b]As reported on RSR. Assumed to mean ^3H_2O.

Table 2.11

Inventory by Trench of H-3 in Shipments $\geq$ 1 Ci

	Amount (Ci)			
Trench	H-3 Listed Separately	Mixture of Isotopes Containing H-3	Estimated H-3 in Mix	Estimated Total H-3
1	305.8	0	0	305.8
2	570.7	130.2	50.9	621.6
7	29.1	2.0	1.6	30.7
11	21.7	11.6	9.4	31.9
14A	684.2	36.0	29.2	713.4
23	162.4	34.8	19.2	181.6
24	286.4	23.3	20.2	306.6
25C	92.2	23.3	18.7	110.9
			Total	2302.5

Table 2.12

Comparison of H-3 Inventories With Total By-Product Material

Trench	Estimated H-3 Inventory (Ci) In Shipments ≥1 Ci	In Shipments <1 Ci	Total H-3	Total By-Product Material (Ci)[a]	Number of Shipments Containing H-3
1	305.8	4.6	310.4	6157.3	66
2	621.6	11.8	633.4	10451.1	209
7	30.7	4.5	35.2	635.8	71
11	31.9	4.5	36.4	1428.1	86
14A	713.4	11.7	725.1	6322.2	193
23	181.6	7.2	188.8	4388.5	119
24	306.6	8.7	315.3	5109.4	126
25C	110.9	3.5	114.4	863.9	67
Totals	2302.5	56.5	2359.0		

[a]From NUS Corporation Report SHEF-C-220 (Draft), Appendix C, October 31, 1979.

2.3.2.1.1 Relative Contribution of Shipments <1 Ci

In the case of the H-3, the amounts in small shipments (<1 Ci) correlates well with the number of shipments, i.e., the trench inventory of H-3 from small shipments is nearly proportional to the number of small shipments. This relationship probably will hold for all the trenches filled in a routine manner which can presumably be attributed to the fact that there was a sufficiently large number of shipments to each trench to provide good statistical sampling.

The number of shipments >1 Ci, however, was generally quite small (Trench 2, with 50, is an obvious exception) and the total amount of H-3 from such shipments depends to a great extent on the particular shippers. One trench may have received one or more shipments >100 Ci, whereas another, such as Trench 11, may have had all shipments <10 Ci. Thus, the relative contribution of small shipments to the total H-3 inventory varies considerably - from 1-1/2% for Trenches 1 and 14A to 12-1/2% for Trenches 7 and 11. However, it is always a relatively small fraction of the total trench inventory, and apparently the amount of H-3 in shipments >1 Ci to a trench provides a good indication of its total H-3 inventory.

2.3.2.2 C-14

In general, the uncertainty associated with the C-14 trench inventories is greater than that for other relevant isotopes because it was more often listed with other isotopes, and no individual amounts given. However, for the four trenches (2, 14A, 23, and 24) which received shipments >1 Ci for which C-14 was listed separately, uncertainties are relatively small, because the amounts of C-14 in mixed low-level waste were small by comparison with the amounts in these large shipments.

The shipments containing >1 Ci for which C-14 was listed separately are given in Table 2.13. Table 2.14 lists the shipments >1 Ci of H-3 which contained C-14 mixed with the H-3, and sometimes other isotopes, but where no breakdown of the amounts by isotope was given. The C-14 amounts listed for these shipments are estimated from information obtained from the shippers, as discussed in Section 2.3.1.2. For the low-activity shipments, this information on the proportion of C-14 in packages of mixed isotopes is still valid for the same shippers. Since H-3 predominated in the mixtures for all the shippers contacted, it was assumed that the same situation applied to most other shippers. To allow for those where it may not have, all the low-activity waste was assumed to have a 4:1 ratio of H-3 to C-14. Allowance was also made for amounts of other isotopes present. The results are given for all the trenches in Table 2.15, along with amounts for large shipments and the estimated total trench inventories.

Table 2.13

Shipments $\geq$1 Ci of C-14 Alone to all Eight Trenches

Trench	Shipper	RSR	Date	Amount (Ci) and Form
2	Mallinckrodt	----	9/10/68	$\leq$6, solid
2	Amersham/Searle	----	2/04/69	7.0, solid
2	Amersham/Searle	4537	10/22/69	2.1, solid
2	Amersham/Searle	3763	3/06/70	3.0, solid
2	Amersham/Searle	3764	7/17/70	2.5, solid
2	Amersham/Searle	3765	10/16/70	2.5, solid
24	Pathfinder Labs, Inc.	03009	11/11/75	$\leq$14, contaminated paper and glass
24	Pathfinder Labs, Inc.	01557	3/30/76	$\leq$4.1, contaminated paper and glass
23	Pathfinder Labs, Inc.	01558	8/03/76	$\leq$6, solid
23	Pathfinder Labs, Inc.	29292	10/26/76	$\leq$6, solid
14A	Nalco Environmental	34414	8/12/77	$\leq$1.5, solid
14A	Pathfinder Labs, Inc.	29541	9/13/77	14, solid
14A	Pathfinder Labs, Inc.	36358	12/19/77	6.0, solid
14A	Pathfinder Labs, Inc.	29306	3/01/78	6.0, solid

Table 2.14

Estimated Amounts of C-14 Mixed With Other Isotopes in Shipments of H-3 ≥1 Ci

Trench	Shipper	RSR	Date	Amount (Ci) and Form
2	Atomic Disposal Co.	----	9/22/69	1.6, solid
2	Miles Laboratories	----	3/19/70	0.4, solid
2	Atomic Disposal Co.	3404,5	3/30/70	0.4, solid
11	Michigan State Univ.	2528	4/09/75	0.2, solid
11	3M Co.	1792	4/23/75	1.5, solid
11	WARF Institute	10574	4/23/75	0.2, liquid
24	Univ. of Wisconsin	12439	10/23/75	0.3, liquid
24	Amersham/Searle Corp.	12460	11/13/75	0.4, liquid
24	Atomic Disposal Co.	20312, etc.	2/16/76	0.6, solid
24	Amersham/Searle Corp.	12461	2/20/76	0.3, solid
25C	University of Wisconsin	12444,5	5/27/76	1.8, mostly solid
25C	Amersham/Searle Corp.	12462	5/28/76	0.7, solid
25C	Amersham/Searle Corp.	12463	6/16/76	0.3, solid
25C	Abbott Laboratories	23650	7/23/76	0.6, solid
23	Amersham/Searle Corp.	31689	9/09/76	0.5, solid
23	3M Co.	20075, etc.	10/05/76	1.1, mostly solid
23	Amersham/Searle Corp.	31692	10/22/76	0.4, solid
23	University of Wisconsin	12447	10/28/76	0.4, liquid
23	Mobay Chemical Corp.	12572	12/02/76	0.7, solid
23	Amersham/Searle Corp.	31693	12/03/76	0.3, solid, water-based radiochemicals
14A	Atomic Disposal co.	36854	9/30/77	0.2, solid
14A	Amersham Corp.[a]	23334	12/12/77	0.7, solid
14A	Amersham Corp.[a]	23335	2/03/78	0.3, solid
14A	Amersham Corp.[a]	51143	3/31/78	0.4, solid

[a]Formerly Amersham Searle Corp.

Table 2.15

Estimated Trench Inventories of C-14

Trench	Amount (Ci) In C-14 Shipments >1 Ci	Estimated Amount in H-3 Shipments >1 Ci	Estimated Amount in All Other Shipments	Estimated Total C-14
1	0	0	1.2	1.2
2	23	2.4	3.1	28.5
7	0	0	0.4	0.4
11	0	1.9	0.6	2.5
14A	27	1.6	1.8	30.4
23	12	3.4	0.9	16.3
24	18	1.6	0.9	20.5
25C	0	3.4	0.5	3.9
			Total	103.7

2.3.2.3 I-129

There were a total of six shipments to four trenches which contained I-129. Information on the shipments is summarized in Table 2.16. All the generators were requested for a description of their waste. The broker, Atomic Disposal Co., is no longer in existence and, therefore, could not be contacted. It is most likely the I-129 in its packages was in the form of sources, as was that in all the other shipments except that of the University of Illinois.

Miles Laboratory, Searle Analytic, and Amersham Corporation personnel sated that the only use for I-129 of which they were aware was as γ-calibration sources, which normally contained a fraction of a μCi each. The amounts in the Miles Laboratory and Amersham Corporation shipments were known (given in the table), and Searle Analytic personnel were certain that their amounts were similar. These μCi amounts of I-129 were contained in >100 mCi packages for all three shippers. The total package activities for the Atomic Disposal Co. packages, on the other hand, were much smaller, ranging from 0.8 to 8 mCi. Thus, it is almost certain that the amounts of I-129 in them were similar to those of the other three shippers, especially since the other isotopes in the mixtures were normally shipped in amounts of the magnitude of the total package activities.

The shipment from the University of Illinois was unlike all the others, both in magnitude and in form. The University Radiation Safety Officer confirmed the amount. (See Memo to File "I-129 Shipments to Trench 2," by D. R. MacKenzie, August 8, 1983, reproduced in Appendix B.) The isotope had been used, not for sources, but for experiments in high energy physics, presumably in targets. The chemical form was probably NaI, the form in which it would almost certainly have been purchased. The Radiation Safety Officer stated that much of the material was unused and was disposed of along with the targets when the experiments were finished.

Table 2.16

Packages Containing I-129 in Trenches 2, 11, 14A, and 23

Trench	Shipper	RSR	Date	Package Number	Other Isotopes in Package	Total Package Activity (mCi)	Amount of I-129 (mCi)
2	University of Illinois	11422	9/17/69	6	Co-57, Te-129m	1.4	1.0
2	Atomic Disposal Co.	4076-8	11/11/70	858	P-32, I-131, C-14, H-3	8	[a]
				878	H-3, I-131, C-14, Tc-99m	2	[a]
11	Miles Laboratory	12102	2/06/75	7	P-32, H-3, I-125, Cs-137, Ba-133	153	0.004
23	Searle Analytic	3688	10/22/76	1	H-3, Ni-63, Co-57 Ba-133, C-14, Sr-90	358	[a]
14A	Atomic Disposal Co.	36876-9	12/01/77	105	I-125, H-3, C-14, S-35	0.8	[a]
14A	Amersham Corp.	23334	12/12/77	11	I-125, Cs-137, Se-75	200	0.0006[b]

[a]Amount not listed separately.
[b]Amount not listed on RSR, but made available by Amersham personnel.

2.3.2.4 Sr-90

Unexpectedly large amounts of Sr-90 were found to have been sent to Trenches 1 and 2 by a single shipper. The amounts shipped to other trenches were relatively small - of a size similar to the amounts of C-14. Tables 2.17 and 2.18 list the shipments >1 Ci to Trenches 1 and 2, respectively, while the shipments >1 Ci to the other trenches are given in Table 2.19. The isotopic distribution in the IIT shipments is not known. For trench inventory purposes, equal distribution has been assumed.

Table 2.17

Sr-90 Shipments ≥1 Ci to Trench 1

Shipper	RSR	Date	Amount (Ci) and Form
3M Co.	----	5/02/67	250, solid
3M Co.	----	5/11/67	55, solid
3M Co.	----	9/14/67	23, solid
3M Co.	----	10/11/67	15, solid
Nuclear Chicago	----	6/06/68	1.1, solid
3M Co.	----	8/06/68	50, solid
ANL special series of shipments			10.8 solid
		Total	404.9

Table 2.18

Sr-90 Shipments ≥1 Ci to Trench 2

Shipper	RSR	Date	Amount (Ci) and Form
3M Co.	----	9/26/68	10.0, solid
3M Co.	----	11/26/68	150, solid
Unknown	----	1/31/69	2.1, solid
3M Co.	----	4/08/69	11.0, solid
3M Co.	----	5/28/69	15, solid
3M Co.	11748	8/07/69	7.0, solid
3M Co.	2136	9/30/69	19.4, solid
3M Co.	11344	12/04/69	74.5, solid
3M Co.	3782	1/22/70	42.2, solid
3M Co.	3720	4/02/70	65.3, solid
3M Co.	3721	6/04/70	30.2, solid
3M Co.	3722	7/30/70	51.0, solid
3M Co.	3730	10/08/70	337, solid
3M Co.	3738	3/18/71	47.3, solid
ANL special series of shipments			134, solid
		Total	996.0

Table 2.19

Sr-90 Shipments ≥1 Ci to Trenches 7, 11, 14A, and 24

Trench	Shipper	RSR	Date	Amount (Ci) and Form
7	3M Co.	11665	6/11/74	2.6, solid
11	University of Notre Dame	2284	4/08/75	1.7, solid
11	Dow Chemical Co.	2281,2	5/01/75	2.0, solid
24	Parkwell Laboratory, Inc.	21379	2/06/76	10.0, solid
14A	Illinois Institute of Technology (IIT)	----	9/13/77	3.0, solid, Sr-90 + Cs-137
14A	IIT	----	10/11/77	13.1, solid, Sr-90 + Cs-137 + Co-60
14A	IIT	----	10/20/77	1.2, solid, Sr-90 + Cs-137
14A	IIT	----	2/06/78	6.1, solid, Sr-90 + Cs-137
14A	IIT	----	2/28/78	2.6, solid, Sr-90 + Cs-137

The special series of shipments from ANL to Trenches 1 and 2, described in Section 2.1.2, contained a considerable amount of Sr-90. The amounts in the two trenches have been estimated in the following way, starting with values for the total curies of 108 in Trench 1 and 1346 in Trench 2. It is known that the shipments contained waste associated with nuclear reactor fuel (e.g, waste resulting from hot cell work with fuel, such as sectioning and examining irradiated fuel assemblies) but the proportion of the total due to such waste is not known. We have assumed that half the waste was associated with irradiated fuel, and therefore contained fission products and activation products (the latter associated with cladding, particularly stainless steel cladding).

It is also known that a lot of this waste had been stored for many years before shipment so that the principal fission products present would be Cs-137 and Sr-90, and the principal activiation product would be Co-60. However, some of the waste was of relatively recent origin, so not all the fuel-related waste can be allocated to Cs-137, Sr-90, and Co-60 since shorter-lived nuclides such as Cs-134, Mn-54, Cr-51, etc., would contribute appreciable fractions of the total curie amount. We have assumed that these latter nuclides contribute half the activity of the fuel-related waste. Thus, the longer-lived Cs-137, Sr-90, and Co-60 are assumed to have contributed one quarter of the total waste in the series shipments. Because fission yields and half-lives for Sr-90 and Cs-137 are roughly the same, these isotopes are assumed to have been present in equal amounts, since no chemical fractionation would have taken place in the production of this type of waste. We have assigned the relative fractions 0.4, 0.4 and 0.2, respectively, to the Cs-137, Sr-90, and Co-60. This leads to estimated amounts of, respectively, 10.8, 10.8, and 5.4 Ci to Trench 1 and 134, 134, and 67 Ci to Trench 2. The values for Sr-90 are included in Tables 2.17 and 2.18.

The estimated trench totals are given in Table 2.24. For Trenches 1 and 2, the Sr-90 amounts are of the order of 100 times as large as the >1 Ci amounts sent to the other trenches. The amounts in shipments <1 Ci sent to the other trenches are small fractions of the amounts in shipments >1 Ci sent to those trenches. For trenches 7, 11, and 14A, the amounts were approximately 0.1 Ci each. Trench 24 had about 0.5 Ci, and Trenches 23 and 25C, which had no large shipments, received 0.24 and 0.04 Ci, respectively, from small shipments. The amounts <1 Ci sent to Trenches 1 and 2 were of the order of a curie, which is trivial compared with the trench totals of hundreds of curies.

2.3.2.5 Cs-137

The same situation exists for Cs-137 as for Sr-90. Over 1000 Ci was shipped to each of Trenches 1 and 2, which is of the order of 100 times the amounts sent to the other trenches, except for Trench 14A which received about 50 Ci. The shipments >1 Ci to Trenches 1 and 2, including the special series shipments from ANL, are given in Tables 2.20 and 2.21, respectively, along with total trench inventories. Table 2.22 lists the shipments >1 Ci to the other trenches, and all the individual trench totals are listed in Table 2.24.

Table 2.20

Non-Fuel Cycle Cs-137 Shipments ≥1 Ci to Trench 1

Shipper	RSR	Date	Amount (Ci) and Form
3M Co.	----	5/02/67	385, solid
3M Co.	----	5/11/67	94.5, solid
3M Co.	867	7/18/67	91.5, solid
3M Co.	----	9/14/67	14, solid
3M Co.	----	10/11/67	10.5, solid
3M Co.	----	12/20/67	21.5, solid
3M Co.	----	1/18/68	12.5, solid
3M Co.	----	3/20/68	33.1, solid
3M Co.	----	4/25/68	14.9, solid
3M Co.	----	6/11/68	153, solid
3M Co.	----	8/06/68	66.2, solid
3M Co.	----	8/15/68	102.5, solid
ANL special series of shipments			10.8,[a] solid
		Total	1010.0

[a]Estimated as described in text, Section 2.3.2.4.

Table 2.21

Non-Fuel Cycle Cs-137 Shipments ≥1 Ci to Trench 2

Shipper	RSR	Date	Amount (Ci) and Form
Amersham/Searle	----	9/09/68	1.1, solid
3M Co.	----	9/26/68	25.0, solid
3M Co.	----	11/26/68	118, solid
3M Co.	----	1/21/69	219.5, solid
Unknown	----	1/31/69	4.3, solid
Amersham/Searle	----	2/04/69	4.4, solid
3M Co.	----	4/08/69	13.5, solid
3M Co.	----	5/28/69	54.0, solid
3M Co.	11748	8/07/69	88, solid
3M Co.	2136	9/30/69	48.4, solid
Amersham/Searle	4537	10/22/69	1.1, solid
3M Co.	11344	12/04/69	23, solid
3M Co.	3782	1/22/70	56, solid
3M Co.	3720	4/02/70	54.9, solid
3M Co.	3721	6/04/70	31.3, solid
3M Co.	3722	7/30/70	109.5, solid
3M Co.	3730	10/08/70	96, solid
Nuclear Chicago	3767	11/06/70	19.7, solid
3M Co.	3731	12/03/70	3.0, solid
Atomic Disposal Co.	4079	3/16/70	3.6, solid
3M Co.	3738	3/18/71	4.4, solid
ANL special series of shipments			134,[a] solid
		Total	1092.7

[a]Estimated as described in text, Section 2.3.2.4.

Table 2.22

Non-Fuel Cycle Cs-137 Shipments ≥1 Ci to Trenches 7, 11, 14A, 23, and 24

Trench	Shipper	RSR	Date	Amount (Ci) and Form
7	3M Co.	11665	6/11/74	2.6, solid
11	Texas Nuclear	0760	4/11/75	5.0, solid
24	3M Co.	20031	9/10/75	1.7, solid
24	3M Co.	20806	9/26/75	2.9, solid
24	Parkwell Laboratory, Inc.	21379	2/06/75	1.0, solid
24	3M Co.	20050	3/04/76	4.2, solid
23	Chicago Metropolitan Sanitation District	03029	10/01/76	1.8, solid, Cs salt
23	3M Co.	20075, etc.	10/05/76	9.0, solid
23	U.S. Air Force, Kelly Airforce Base	31982	10/21/76	13.6, solid, Cs-137+ Kr-85 + Co-60
23	Cleveland-Cliffs Iron Co.	34415	12/02/76	3.7, solid sources
14A	Kay-Ray	1710	6/15/77	5.0, solid
14A	Dow Chemical	22527, etc.	8/16/77	4.0, solid
14A	Illinois Institute of Technology	----	9/13/77, etc.[a]	12.9, solid, Cs-137+ Sr-90
14A	Illinois Institute of Technology	----	10/11/77	13.1, solid, Cs-137+ Sr-90 + Co-60
14A	University of Wisconsin	36729,30	10/13/77	<16.4, solid
14A	3M Co.	20632, etc.	11/16/77	11.1, 3M brand radiating microspheres
14A	Kay-Ray	02026	11/16/77	10, solid

[a]Same series of shipments listed separately in Table 2.19.

As with Sr-90, the amounts of Cs-137 in shipments <1 Ci to Trenches 1 and 2 would be trivial and have not been determined. In the case of Cs-137, the amounts in shipments <1 Ci sent to the other trenches are insignificant compared to those in the fuel cycle waste. For example, Trenches 7, 11, and 25C each contained only about 10 mCi or less. Trench 23 contained approximately 1.5 Ci, still <1% of the fuel cycle contribution. The only trench which received amounts >1% of the estimated fuel cycle amount in shipments <1 Ci was Trench 14A, with approximately 3.3 Ci.

2.3.2.6 Co-60

The shipments of Co-60 ≥1 Ci to all trenches are given in Table 2.23. The total amounts for each trench are listed in Table 2.2.4. No trenches received unexpectedly large amounts of Co-60 from non-fuel cycle shippers, but the amounts sent to Trenches 1 and 2 were roughly 10 times that sent to any other trench. The largest single shipment was one containing 80 Ci, shipped by the University of Wisconsin. Both this and their 4.4 Ci shipment of December 12, 1967 probably consisted of commercial irradiators, since it was known that that was the nature of the 4.4 Ci source. The special series shipments from ANL are responsible for the large Trench 2 total. As with Cs-137, the amounts of Co-60 in shipments <1 Ci were generally small, and proportionately even smaller compared to the fuel cycle contribution, since the latter is larger for Co-60 than for Cs-137. Three of the trenches received <100 mCi in shipments <1 Ci; the largest amount from the small shipments, approximately 2.3 Ci, went to Trench 14A. In fact, the total non-fuel cycle amounts are insignificant, except for that in Trench 1 with its approximately 90 Ci non-fuel cycle contribution.

Table 2.23

Non-Fuel Cycle Co-60 Shipments ≥1 Ci to All Trenches

Trench	Shipper	RSR	Date	Amount (Ci) and Form
1	3M Co,	-----	7/18/67	4.0, solid
1	University of Wisconsin	-----	10/10/67	80, solid source
1	University of Wisconsin	-----	12/27/67	4.4, solid source
2	Nuclear Chicago Corp.	3767	11/06/70	4.3, solid
2	Nuclear Chicago Corp.	3750	12/04/70	1.4, solid
24	Ford Motor Co.	23078	4/06/76	1.6, solid, in lead holders
23	U.S. Air Force, Kelly Air Force Base	31982	10/21/76	13.6, solid, Cs-137+ Kr-85 + Co-60
14A	Dow Chemical	22527, etc.	8/16/77	2.3, solid
14A	Kay-Ray	35932	9/08/77	4.0, solid
14A	Neutron Products, Inc.	18735	10/07/77	1.2, solid (wood, steel, dirt, cement)
14A	Illinois Institute of Technology	-----	10/11/77	13.1, solid, Cs-137+ Co-60 + Sr-90
14A	Illinois Institute of Technology	-----	2/15/78	1.0, solid
1	ANL special series of shipments			5.4,[a] solid
2	ANL special series of shipments			67,[a] solid

[a]Estimated as described in text, Section 2.3.2.4.

Table 2.24

Estimated Trench Non-Fuel Cycle Inventories of Sr-90, Cs-137, and Co-60

Trench	Amount (Ci)		
	Sr-90	Cs-137	Co-60
1	406	1010	97
2	996	1093	75
7	3	3	<1
11	4	5	<1
14A	11	60	15
23	<1	21	5
24	11	10	2
25C	<1	<1	<1

2.3.2.7 Transuranic (TRU) Isotopes

The TRU elements shipped in significant amounts were Pu and Am. Only very small amounts of Np-237 and Cf-252 were observed - approximately 0.1 mCi for Np-237 in five shipments, and 3.6 mCi for Cf-252 in six shipments. Pu was usually in the form of its mass 239 to 241 isotopes, but there were several shipments containing Pu-238. The largest amounts of Pu came in shipments (from Kerr McGee Corp.) we have classed as fuel cycle, but they are included with the non-fuel cycle inventory in order to have the data on TRU waste all in one place. Am-241 was shipped in significant amounts, but much smaller than Pu. The shipments of the TRU isotopes >1 Ci are listed in Tables 2.25 and 2.26. The amounts in shipments <1 Ci were very small: approximately 100 mCi of Am-241 went to each of Trenches 23, 24, and 25C; approximately 100 mCi of Pu went to Trench 2, and a few mCi or less went to each of the other trenches.

The curie amounts of Pu in the ANL shipments were estimated from the weights given on the RSRs (in parentheses in the table). It was assumed that the Pu-241 concentration was 0.1 weight percent. This could be high for Pu which originated in lightly irradiated fuel, but would be low for highly irradiated fuel. It is known that the ANL special series of shipments, of which this waste was a part, contained waste from hot cell examination of EBR-1 and EBR-2 fuel. The portions of this waste containing Pu fuel would undoubtedly be higher than 0.1 weight percent in Pu-241. However, since there is no record of relative amounts of the various wastes in the special series of shipments, we have no way to make a more precise estimate than we have done. If the average Pu-241 content was greater than 0.1%, the curie level from ANL would be greater than the amounts in the table. However, weights listed by ANL were usually given as an upper limit so it is unlikely that they shipped much greater curie amounts than we have estimated. In any case, the total ANL amount is small compared with the total amount shipped by Kerr McGee.

For the Kerr-McGee shipments, the Pu was reported as Pu-239 and 241, and the weight was given in the column for SNM. Only odd Pu isotopes are fissile, so even Isotopes are not counted as SNM, but obviously the material would have contained appreciable Pu-240, and small amounts of Pu-238 and Pu-242. For two of the shipments (those in Table 2.25 with the weights in parentheses), the curie amounts were also given. This allows us to calculate the Pu-241 contents to a fairly close approximation. That for shipment 4375 to Trench 2 was approximately 1.7 weight percent, and that for shipment 2350 to Trench 7 was approximately 1.9 weight percent. Thus, it appears that the isotopic content of the Pu was roughly the same for all the shipments, and we have calculated the curie amounts using the information from those two shipments where both weight and curie amount were given.

The inventory information for the transuranic elements is summarized in Table 2.27. Shipments which contributed <1 Ci are not included since their total amounts were at most about 100 mCi for any trench. Total trench TRU waste amounts are given, as well as amounts of Pu and Am separately. The total activity of TRU waste (largely due to Pu-241) is approximately 2000 Ci.

The Pu-241 component (roughly 1850 Ci) is a relatively large amount in the near term, and in 100 years, after most of it has decayed, it will have grown into ~65 Ci of Am-241, which is still an appreciable quantity for the long term.

Table 2.25

Shipments of Pu-238 and Pu-239,241 ≥1 Ci

Trench	Shipper	RSR	Date	Amount (Ci) and Form
2	Argonne National Laboratory (ANL)[a]	----	4/30/69	23.4 (120 g), solid
2	ANL	4193,-6	6/22/70	21.8 (113 g), solid
2	ANL	4147, etc.	6/23/70	97 (500 g), solid
2	ANL	4293	7/07/70	5.8 (30 g), solid
2	ANL	4292	7/08/70	3.7 (19 g), solid
2	Kerr McGee Nuclear Corp.[b]	4372	7/21/70	105, solid
2	Kerr McGee	4367	9/11/70	59, solid
2	Kerr McGee	4368,9	10/22/70	57, solid
2	Kerr McGee	4370	11/18/70	88, solid
2	Kerr McGee	4373	12/29/70	122, solid
2	Kerr McGee	4374	2/04/71	57, solid
2	Kerr McGee	4375	3/04/71	82 (40.8 g),[c] solid
7	Kerr McGee	11117	3/26/74	63, solid
7	Kerr McGee	11118	3/28/74	108, solid
7	Kerr McGee	11120	4/16/74	44, solid
7	Kerr McGee	2781	4/30/74	78, solid
7	Kerr McGee	2783	5/21/74	111, solid
7	Kerr McGee	2784	5/30/74	123, solid
7	Kerr McGee	2350	6/12/74	132 (60.0 g),[c] solid
7	Kerr McGee	2785	6/14/74	132, solid
11	U.S. Army (Charlestown)	2499	3/20/75	2.5, in Pu-Be source
11	Kerr McGee	07135,6	5/05/75	432, solid
24	Parkwell Laboratory	21379	2/06/76	3.0 (Pu-238), solid

[a]Curie amounts estimated as described in text, Section 2.3.2.7.
[b]Curie amounts calculated on the basis of RSRs 4375 and 2350.
[c]Both weight and curie amount given on RSR.

Table 2.26

Shipments of Am-241 $\geq$1 Ci

Trench	Shipper	RSR	Date	Amount (Ci) and Form
2	3M Co.	----	9/26/68	1.5, solid
11	Gamma Industries	1426	3/17/75	5.0, solid
24	USAF (Marshal Space Flight Center)	1354	9/11/75	5.0, solid
24	Gamma Industries	13652	10/21/75	7.0, Am oxide
24	Parkwell Laboratory, Inc.	21379	2/06/76	28.6, solid
24	Gamma Industries	00465	2/25/76	3.0, solid
24	Gamma Industries	9320	4/20/76	$\leq$1.0, solid
24	Gamma Industries	23037	5/04/76	$\geq$2.6, AmO_2
24	Kay-Ray	01703	5/14/76	1.0, Am-Be sources

Table 2.27

Trench Inventories of Significant Amounts of TRU Waste

Element	Amount (Ci)				
	Trench 2	Trench 7	Trench 11	Trench 24	All Trenches
Pu	722	791	435	3.0	1951
Am	1.5	0	5.0	48.2	54.7
Total TRU	723.5	791	440	51.2	2005.7

Acceptance of TRU waste was discontinued by 1976 at all LLW burial sites except Hanford.[6] According to a NUREG report (Reference 3, p.3-38), Sheffield stopped accepting TRU waste in 1975. However, Parkwell Laboratory, Inc., Gamma Industries, and Kay-Ray all shipped appreciable quantities in 1976. No records of shipments of waste which could be classed as TRU waste were observed after the Kay-Ray shipment of May 14, 1976, and on the RSR of a shipment by BRK Electronics on November 19, 1976 is a note to the effect that the shipper had been contacted to ensure that the Am-241 contained in the shipment was below the 10 nCi/g level which defined TRU waste.

2.3.2.8 Long-Lived β-Emitters

Several long-lived ($t_{1/2} > 10^4$ yr) pure β-emitters are included in the list of isotopes of concern in the management of low-level waste (Table 3.8 of Reference 6). They are of concern not only because of their very long half-lives, but also because of the difficulty in measuring and monitoring them due to their lack of γ-rays. The isotopes in this category are:

Ni-59, $t_{1/2} = 8 \times 10^4$ yr

Nb-94, $t_{1/2} = 2.0 \times 10^4$ yr

Tc-99, $t_{1/2} = 2.1 \times 10^5$ yr

Ni-63, with $t_{1/2} = 100$ yr, is included in the table, and we would also include Cl-36 with $t_{1/2} = 3.0 \times 10^5$ yr.

We have already discussed I-129 (Section 2.3.2.3), a β,γ-emitter whose half-life is 1.6×10^7 yr. Only six shipments containing I-129 were noted for the eight trenches of this study, and all except one of these contained only a few μCi or less. The one exception contained 1 mCi. The long-lived pure β-emitters turn out to have been shipped with a similar frequency, when they were shipped at all. Nb-94 and Ni-59 were not shipped to any of the trenches whose records were examined. Ni-63 was received in all eight trenches, but only in mCi amounts (usually 40-50 mCi to a trench, but approximately 200 mCi in Trench 24). Tc-99 is recorded as having been present in a number of shipments from medical institutions, but this is almost certainly incorrect reporting of Tc-99m ($t_{1/2} = 6$ h), since the other isotopes these institutions shipped were short-lived isotopes such as I-131 ($t_{1/2} = 8$ d), and Tc-99m is used regularly in medical applications. Tc-99 was definitely shipped by 3M Co., but only in mCi amounts. The amounts which could be positively identified as Tc-99 were about 10-20 mCi per trench, except for Trench 2 which contained about 300 mCi. Experience with Cl-36 was similar, the total amount shipped being several hundred mCi.

These isotopes were buried in such small amounts that development of scenarios for their probable release rates is considered to be unproductive. An uncommon isotope, the positron emitter, Al-26 ($t_{1/2} = 7.3 \times 10^5$ yr) occurred in one shipment in the relatively large amount of 6 Ci. This isotope was contained in highly irradiated aluminum fuel rod ends. Thus, its release to the trench would be extremely slow due to the very low solubility of aluminum and/or its Al_2O_3 film under trench conditions. As with the long-lived pure β-emitters, efforts to develop a release scenario would presumably not be worthwhile.

2.3.3 Fuel Cycle Inventory

2.3.3.1 Definition of Fuel Cycle Waste

Fuel cycle waste, for purposes of this report, consists of waste connected with the operation of commercial nuclear power plants. This includes waste associated with fuel fabrication, which was largely source material or special nuclear material (SNM). The bulk of the by-product material in the fuel cycle category obviously came from the power plants, not the fuel fabrication plants.

It was assumed that waste from reactors connected with defense work would be handled by DOE and would not be shipped to commercial burial sites. In principle, research reactors at institutions and national laboratories produce fuel cycle waste, and it can be argued that such waste should be included with that generated by commercial power reactors. However, it is difficult, in fact often not possible, to distinguish between waste produced at the reactors and that generated in other departments of the institutions. In any case, the amount involved is small compared to total fuel cycle waste, so we have classed all waste from institutions and national laboratories as non-fuel cycle. The distinction has no significance in terms of estimating release rates to the trenches, since source terms for the trenches consist of all the waste that has been buried, including the small amount we have classed as simply "other" because we were not sure how to classify it.

2.3.3.2 Trench Inventory of By-product Material

As mentioned in Section 2.3.2.7, Pu from all shipments is considered in that section, whether shipments were classed as fuel cycle, non-fuel cycle or other, since origin is not of concern as far as the inventory is concerned. Thus, in this section, we consider only the by-product material in the fuel cycle shipments.

The information on fuel cycle shipments is summarized in Table 2.28. This gives a breakdown by trench of the numbers of shipments and percentages of total fuel cycle waste sent by the largest generators, where they are known, and by the brokers Atcor and Anefco where the origin of the waste was not given. The criterion for choice of generators was contribution of $\geq$10% of the inventory to at least one trench. It should be noted that California Nuclear, Inc. (CNI) shipped the bulk of the waste to Trench 1. CNI is not included in the body of the table because it was the operator of the Sheffield site at the time, but had no further part in operations, and was thus neither a generator, nor a shipper in the usual sense.

Table 2.28

Summary of Information on Fuel Cycle Waste Shipments

	Trench Number							
	1[a]	2	7	11	14A	23	24	25C
Number of Shipments								
All Fuel Cycle	27	83	302	355	823	474	972	293
Commonwealth Edison	7	40	104	241	554	311	676	106
ANEFCO	0	0	161	8	0	13	212	164
ATCOR	2	33	22	69	64	35	26	2
Consolidated Edison	2	0	0	0	22	25	1	1
Northern States Power[b]	0	0	5	12	23	0	25	4
Nebraska Public Power District	0	0	0	4	25	15	27	7
Chem Nuclear Systems[b]	0	0	0	4	4	9	0	1
Radioactivity								
Total Curies	2299	1719	241	1276	4701	4204	4293	728
Percent of Total From:								
Commonwealth Edison	11.0	0.9	41.1	40.0	74.8	71.5	62.0	61.3
ANEFCO	0	0	40.7	<0.1	0	<0.1	4.4	14.2
ATCOR	0.6	97.3	8.7	10.7	3.1	2.8	4.4	1.6
Consolidated Edison	0.6	0	0	0	9.6	11.3	0.9	1.9
Northern States Power[b]	0	0	10.0	37.2	4.8	0	24.1	2.9
Nebraska Public Power District	0	0	0	0.7	2.7	0.4	4.0	16.5
Chem Nuclear Systems[b]	0	0	0	10.8	2.1	10.7	0	<0.1

[a]The largest fuel cycle shipper to Trench 1 was California Nuclear, Inc., predecessor of NECO at Sheffield, contributing 85% in 13 large shipments. Origin of this waste is not known.
[b]Eight large shipments from Chem Nuclear Systems to Trench 11 and six to Trench 24 originated at the Monticello reactor and are therefore listed with Northern States Power Company, operator of the reactor.

2.3.3.3 H-3 Component of Fuel Cycle Waste

In general, H-3 was not given on RSRs as a constituent of the fuel cycle waste. From contacts with power plant personnel, it appears that power plants were not analyzing their waste for H-3 during the period of Sheffield's operation. Of the more than 3000 fuel cycle RSRs covered by our eight trenches, only one mentioned H-3. This one, for a Commonwealth Edison shipment of June 23, 1976 to Trench 25C, listed the major isotopes as MFP and H-3, but did not give a separate amount for H-3. An estimate of the fuel cycle contribution to the Sheffield site H-3 inventory was made in the Draft EIS prepared by NUS Corp.(1) This was based, in part, on data contained in an EPA report on radioactivity released from nuclear power plants in the period 1973 through 1977.(7) The estimate arrived at for the whole site was 159 Ci. This estimate used the amounts of H-3 in liquid effluent to establish H-3 concentrations in the liquid waste streams of PWRs, which tended to be an order of magnitude higher than those of BWRs. These concentrations, of the order of 10^{-2} μCi/mL, would not change during production of evaporator bottoms or concentrate. Thus, amounts of H-3 appearing in the waste could be estimated from the volumes of concentrate disposed of.

The calculation used an average value of the H-3 concentration based on the experience of PWRs, several of which did not contribute an appreciable fraction of the total fuel cycle waste. It also assumed that the amounts of waste shipped to Sheffield by power plants was proportional to their numbers. Thus, the NUS report states that about as many PWRs as BWRs shipped waste to Sheffield, and assumed that the amounts from each type were equal. This is not the case for the trenches covered by our study, where Commonwealth Edison BWRs alone shipped 3/4 of the fuel cycle waste to two of the three largest trenches (in terms of amount of fuel cycle activity buried). Approximately 3/4 of the fuel cycle waste in five of our eight trenches was from BWRs. This fraction may be even higher when waste shipped by brokers is taken into account, but we do not know where their shipments originated. Essentially all the fuel cycle waste in Trench 2 (9% of the total in the eight trenches studied) was from Consolidated Eidson's Indian Point PWRs. It is interesting to note that the H-3 concentration in their liquid waste[7] was an order of magnitude lower than the value used by NUS for PWRs.

Because of the method of choosing an average value for H-3 concentration in PWR liquid streams, and the assumption of equal amounts of PWR waste and BWR waste, the H-3 site inventory estimated by NUS is undoubtedly high. If the rest of the site had the same mix of BWR and PWR waste as did our eight trenches, the inventory would be down by at least a factor of 2. Thus, the value of site H-3 inventory which we have chosen for use in estimating our trench H-3 inventories is 80 Ci.

Using the values for trench total radioactivity given in Table 1.1, taken from Tables C-41 and C-42 of the NUS Corp. report,[1] one finds that the eight trenches assigned to this study received slightly more than half the total waste buried at Sheffield. Since our trenches cover the whole operating life of the site (including the first and last trenches in operation), it is reasonable to assume that they contain roughly half the fuel cycle waste, as well as half the total waste. Thus, we have taken a value of 40 Ci for the fuel cycle H-3 in these trenches. Assuming that this fuel cycle H-3 is distributed among the trenches in proportion to the amounts of their fuel cycle waste, the amounts of H-3 due to the fuel cycle waste can be estimated for each trench. These amounts are given in Table 2.29, along with the estimated non-fuel cycle H-3 inventories.

Analyses of radioactive wastes from a number of power plants have been performed by Science Applications, Inc. Results for relevant plants were made available to BNL by NRC*. They consist simply of lists of isotopes contained in the waste, with their concentrations in μCi/g. The dates of sampling and analysis are given, and the type of waste (e.g., filter sludge, evaporator bottoms, or resin), but no information is included on analytical methods. Roughly half the analyses included values for tritium, along with the standard activation product and fission product results. Of the tritium

*Attachment to letter of S. Z. Jones of NRC to W. Y. Kato of BNL, April 2, 1984.

Table 2.29

Estimated Amounts of H-3 in Fuel Cycle Shipments

Trench	Amount of Fuel Cycle Waste in Trench (Ci)	Amount as % of Total	Estimated Amount of Fuel Cycle H-3 in Trench (Ci)	Estimated Amount of Non-Fuel Cycle H-3 in Trench (Ci)[a]
1	2299	11.8	4.7	310
2	1719	8.8	3.5	633
7	241	1.2	0.5	35
11	1276	6.6	2.6	36
14A	4701	24.2	9.7	725
23	4204	21.6	8.6	189
24	4293	22.1	8.8	315
25C	728	3.7	1.5	114

[a]Taken from Table 2.12.

results, more than half were given only as upper limits. Two of the 25 results not given as upper limits showed a H-3 content >1% of the total activity; the remainder were in the range of 0.005 to 0.5% and mostly of the order of 0.1%. The results given as upper limits were generally considerably <0.1%.

These results indicate that power plant wastes tend to contain a few tenths of a percent of tritium at most, and usually ca 0.1% or less. Assuming this reasoning can be applied to the plants shipping waste to Sheffield (which is, in fact, the case for the few large shippers to Sheffield included in the Science Applications, Inc. compilation), it is concluded that the fuel cycle tritium in the 8 trenches of this study was probably about 20 Ci, which is even less than the value of 40 Ci we had arrived at. In any case, the fuel cycle contribution is so much smaller than the non-fuel cycle that it can be neglected for purposes of release scenario development.

2.3.3.4 Estimation of Amounts of Relevant Isotopes

In general, no breakdown into amounts of the various isotopes can be made for fuel cycle waste from the data given on the RSRs. In Section 2.3.3.2, the trench inventories were given in terms of total activity, but this tells us nothing about what amounts of relevant isotopes were in the waste. For probably half the waste, the designation MFP was used, so for that waste we are not told which isotopes were present, let alone what the individual amounts were. Another standard listing in the column for "major isotopes" was "Co-60 and Cs-137," again with no individual amounts. Several power plants gave a complete breakdown into amounts of all isotopes present in more than trace quantity. The only major shipper listed in Table 2.27 to do this was the Nebraska Public Power District (NPPD). Their data are in line with those of the other plants which gave such a breakdown.

A detailed examination was made of the fuel cycle waste shipped to Trench 24, which had one of the largest fuel cycle waste inventories, and whose ratio of fuel cycle to non-fuel cycle waste was also one of the highest. Results of the examination were helpful not only in estimating amounts of relevant isotopes, but also in developing estimates of their release rates. Data on the RSRs from NPPD contributed greatly to an understanding of the problems connected with determination of the isotopic composition of fuel cycle waste. Two examples of the detail on their RSRs are given in Table 2.30, one for a batch of waste in which no fission products were reported and one for a batch containing the fission products Cs-137 and I-131. I-131 and Ag-110m were seldom reported and are disregarded in our estimations. Mn-54 was usually relatively abundant, as in these examples, but sometimes was not reported. Regardless of differences between different batches of waste, the following pattern is apparent:

- Activation products predominated over fission products.
- The activation products with half-lives <5 yrs predominated over Co-60 ($t_{1/2}$ = 5.3 yr).
- Cs-137, when reported, was generally around 20% of the Co-60, occasionally dropping to as low as 10%.

Also given in Table 2.30 are the total amounts of the different isotopes in the 27 NPPD shipments to Trench 24. It is seen that, on average, the Cs-137 content of the overall waste was only a little over 10% of the Co-60. The Co-60, in turn, contributed only about 20% of the total activity. These results illustrate the point that the RSRs for the bulk of the fuel cycle waste tell essentially nothing about its isotopic composition. The results also provide precise values for amounts of relevant isotopes in NPPD shipments to Trench 24, and these values have been used as the basis for estimating NPPD's contributions of Cs-137 and Co-60 to the other trenches.

Table 2.30

Isotopic Composition of Two Batches of NPPD Waste in Trench 24

Isotope	Amount (mCi) for Batch in Shipment 10327	Amount (mCi) for Batch in Shipment 21977	Total Amount (Ci) in Trench 24
Cr-51	25.2	246.0	78.7
Mn-54	212.0	122.0	28.2
Co-58	158.4	103.0	39.6
Fe-59	10.0		
Co-60	32.4	117.0	39.1
Zn-65	36.0	10.5	3.8
Zr,Nb-95	16.1	13.1	8.1
Ag-110m	2.8		
I-131		88.0	
Cs-137		23.3	4.2

Since there are no comparable data available for most of the fuel cycle waste, the major shippers were contacted to obtain information on which a reasonable estimate of isotopic composition could be based. All the power plants contacted except Monticello (the source of most of Northern States Power's waste) and the Commonwealth Edison plants, Dresden and Quad Cities, reported that their waste contained mostly activation products, usually more than 90%. Monticello listed Cs-134 and Cs-137 for the major isotopes in most of its shipments to Trench 24. For the period when it was shipping waste to the trenches covered by this study, it had relatively high amounts of fission products due to fuel element leaks. Monticello personnel estimated that, on average, Cs-137 and Co-60 contents were nearly equal. Both isotopes represented a major part of their respective fission product and activation product categories, so we have assigned them each 1/3 of the total Monticello contribution.

Commonwealth Edison constitutes a special case since it contributed well over half the fuel cycle waste to the 8 trenches covered by this study, and almost 70% to the 3 largest trenches, including Trench 24 (see Table 2.28). Almost all of this waste was shipped in a little over 3 years, during the period December 1974 to April 1978. In that period both Quad Cities and Dresden were experiencing problems with leaking fuel assemblies, in one unit at Dresden, and in both units at Quad Cities. Trench 24, which operated for almost a year during this time (June 1975 to May 1976), received approximately three times the curie amount from Quad Cities that it did from Dresden. There is no reason to expect that the proportions changed significantly during the remaining time Sheffield was operating, so a value of 3 has been assigned to the Quad Cities/Dresden curie amount ratio. The SAI analytical results mentioned in the last section (2.3.3.3)* contained analyses for 4 Quad Cities waste samples taken in April 1978, the month that Sheffield closed. It was not possible to obtain detailed results for the remainder of the period back to December 1974. However, Quad Cities personnel felt that the SAI results were representative of the entire period. Thus, based on these analyses, assignments of 0.27 and 0.42 have been made for the curie fractions of the waste due to Co-60 and Cs-137 respectively. Analytical results provided by Dresden led to assignment of the values 0.31 and 0.05 for the curie fractions of the Dresden waste due to Co-60 and Cs-137 respectively. These assignments were used for all trenches, and yield the results given in Table 2.31.

For the remainder of the fuel cycle waste, we have assigned a ratio of 10:1 for activation products to fission products, with Co-60 constituting one third of the activation product activity and Cs-137 two-thirds of the fission product activity. These values are, of course, only estimates, based on the information obtained from the shippers to the effect that the Cs-137 content was normally considerably higher than Cs-134, the only other fission product of consequence, and that Co-60 was always one of the principal activation products, but several others (such as Mn-54, Cr-51, and Co-58) were normally

*Attachment to letter of S. Z. Jones of NRC to W. Y. Kato of BNL, April 2, 1984.

Table 2.31

Estimated Amounts of Cs-137 and Co-60 in Fuel Cycle Shipments

	Amount (Ci)									
	Monticello		NPPD		Commonwealth Edison		Remainder		All Shipments	
Trench	Co-60	Cs-137	Co-60	Cs-137	Co-60	Cs-137	Co-60	Cs-137	Co-60	Cs-137
1	0	0	0	0	72	83	620	124	692	207
2	0	0	0	0	6	7	514	103	520	137
7	8	8	0	0	28	32	36	7	72	47
11	158	158	2	0.2	146	167	83	17	389	342
14A	107	107	25	3	1008	1153	249	50	1385	1313
23	150	150	3	0.3	859	982	361	72	1373	1204
24	337	337	39	4	763	871	126	25	1265	1237
25C	0.5	0.5	23	3	128	146	43	9	194	158

present in appreciable proportions. It is quite likely the values, given in Table 2.31, are within a factor of two of the actual amounts present in the waste, assuming the curie amounts reported by the shippers were accurate.

2.3.3.5 Sr-90 Component of Fuel Cycle Waste

Sr-90 was listed as a waste constitutent on only a half dozen of the more than 3000 fuel cycle RSRs we examined, and on those no specific curie amounts were given. Most commercial nuclear power plants analyzed their waste for Sr-90 only occasionally, since the analysis is inconvenient and the amounts found were invariably low.[8] From the information available on Sr-90 content of fuel cycle waste, it was obvious that amounts would be small compared with the non-fuel cycle Sr-90 in Trenches 1 and 2. However, it was not obvious how fuel cycle and non-fuel cycle amounts would compare in the other trenches.

We have estimated the Sr-90 content of the fuel cycle waste in the following manner. Results of a survey on resin usage at commercial power plants[8] indicated that the Sr-90 level in resin wastes was of the order of 1% of the Cs-137. This should hold for liquid waste streams as well (such as that leading to concentrate), since the resins would pick up the trace levels of both Cs and Sr efficiently and fractionation should not occur. Several plants responding to the survey gave estimates well under 1% of the Cs-137, and one of these (a BWR) used a value of 1% routinely, since they considered it an upper limit which was never reached. Only one plant (a PWR) reported a Sr-90 content >1% of the Cs-137. This plant, whose one analysis obtained in the survey was 2.5%, did not ship waste to the trenches covered in this study as far as is known. Even if the Sr-90 content of PWR waste tends to be higher than that of BWR waste, the preponderance of BWR waste in the trenches of concern supports the position that overall the Sr-90 content of the waste would have been <1% of the Cs-137. Contacts with the utilities lead to the same conclusion. Thus, as a conservative estimate, we have taken the Sr-90 content

of the fuel cycle waste to be 1% of the Cs-137. Using the Cs-137 amounts estimated in the previous section (2.3.3.4) leads to the Sr-90 amounts given in Table 2.32.

Table 2.32

Estimated Amounts of Sr-90 in Fuel Cycle Shipments

Trench	Amount (Ci)	
	Fuel Cycle	Non-Fuel Cycle[a]
1	2.0	406
2	1.4	996
7	0.5	2.7
11	3.4	3.9
14A	13.1	11.2
23	12.0	0.2
24	12.4	10.6
25C	1.6	0.1

[a]See Table 2.24.

2.4 Evaluation of Inventory Information

2.4.1 Comparison of Fuel Cycle and Non-Fuel Cycle Wastes

In terms both of volume and activity, fuel cycle waste constituted a far greater proportion of the total waste than did non-fuel cycle for the portion of the site covered in this study. In the case of individual trenches, the fuel cycle contribution was always greater, and usually appreciably greater, than non-fuel cycle, except for Trenches 1 and 2, where it was considerably smaller. In Table 2.33, the comparison is made, in three different ways for each trench; first in terms of the numbers of shipments, then in terms of the total by-product material radioactivity, and finally in terms of the estimated activity of relevant isotopes.

It is apparent from the data on number of shipments that fuel cycle waste did not start coming to Sheffield in large amounts until after at least the first four years of operation (the period when Trenches 1 and 2 were being filled). The ratios of amounts of fuel cycle to non-fuel cycle do not change greatly whether based on total radioactivity or amounts of relevant isotopes. This presumably reflects the fact that the amounts of the short half-life isotopes (of no concern to this study) in the non-fuel cycle waste maintained approximately the same ratio with respect to relevant isotope activities from trench to trench.

Table 2.33

Comparison of Amounts of Fuel Cycle and Non-Fuel Cycle Waste

	Trench							
	1	2	7	11	14A	23	24	25C
Number of Shipments[a]								
Fuel Cycle	27	83	302	355	823	474	972	293
Non-Fuel Cycle	245	453	113	167	302	167	67	80
Ratio $\frac{\text{Fuel Cycle}}{\text{Non-Fuel Cycle}}$	0.11	0.18	2.7	2.1	2.7	2.8	14.5	3.7
Total Radioactivity[b]								
Fuel Cycle (Ci)	2299	1719	241	1276	4701	4204	4293	728
Non-Fuel Cycle (Ci)	2802	8255	197	166	1433	363	707	145
Ratio $\frac{\text{Fuel Cycle}}{\text{Non-Fuel Cycle}}$	0.82	0.21	1.2	7.7	3.3	11.6	6.1	5.0
Estimated Relevant Isotope Activity[c]								
Fuel Cycle (Ci)	900	657	120	731	2698	2577	2502	302
Non-Fuel Cycle (Ci)	1820	2613	42	48	814	229	357	118
Ratio $\frac{\text{Fuel Cycle}}{\text{Non-Fuel Cycle}}$	0.49	0.25	2.8	15.2	3.3	11.2	7.0	2.6

[a]Based on the shipments selected for purposes of this report.
[b]Based on shipment total radioactivity, so includes all by-product material such as isotopes of short half-life, waste designated MFP, etc.
[c]Based on package radioactivity. Does not include packages listed as containing MFP, except for ANL special series of shipments.

2.4.1.1 H-3

Estimated contributions of the fuel cycle waste to the H-3 trench inventories were given in Table 2.29, along with the non-fuel cycle contributions for comparison. The fuel cycle component was 1.7% of the total H-3. For most of the trenches, the amount was close to 1%, but for Trenches 11, 23, and 24, it was 7.2%, 4.6%, and 2.8%, respectively. As pointed out in Section 2.3.3.3, on the basis of analytical results provided us by NRC, it is probable that the actual fuel cycle component of the total buried H-3 was less than indicated by the values given in Table 2.29. Thus fuel cycle contributions to the H-3 inventory were probably <1% for most trenches and <5% even for Trench 11, so that they can be neglected for purposes of estimating H-3 release rates.

2.4.1.2 Sr-90

Because of the biological sensitivity of Sr-90 and the large amount buried at Sheffield, the inventory information on this isotope is important. The estimated fuel cycle Sr-90 contributions to trench inventories were given in Table 2.32, together with the non-fuel cycle contributions for comparison. Due to the large amounts of non-fuel cycle Sr-90 in Trenches 1 and 2, the fuel cycle Sr-90 contribution to those trenches is insignificant. However, its contribution to the other trenches is appreciable, ranging from 16% for Trench 7 to 98% for Trench 23.

Table 4.5, Continued

List of H-3 Packages in Shipments >1 Ci Which Require Special Consideration

Trench	Shipper	RSR or Date	Curies in Shipment	Description of Waste
		T_2/HT Gas in Glass Bulb		
1	Abbott Laboratories	743 6/16/67	13.5 4.5	4 sealed glass bulbs contained in metal pipes closed with threaded caps, no secondary containers
2	Michigan State	3548	25	Sealed glass cylinder embedded in concrete, shipped in drum
14A	University of Illinois	50986	5	5 sealed glass bulbs in iron pipe capped at both ends; shipped in single drum
		Liquids[b]		
7	Mallinckrodt Nuclear	3/04/74	9.6	HTO in 10-mL glass vials with rubber stoppers packed in sorbent in 3 fiberboard drums
25C	University of Missouri	21758	8.0	Toluene solution (solidified at Sheffield)
	Marshfield Medical Foundation	21778	3.5	Alcohol solution (solidified at Sheffield)

[a]A number of shipments contained smaller amounts (but >1 Ci) of activity in general waste. These are included in the calculations carried out in Section 4.4.5.

[b]A number of shippers of amounts >1 Ci sent amounts of liquid waste smaller than those listed here. These amounts are considered on a trench-by-trench basis in the calculations (Section 4.4.9).

4.4.1 Targets

4.4.1.1 Targets Shipped by BNL

BNL vault 3-137 contained several tritiated titanium targets (activity 21.1 Ci) in double glass jars which were then placed in polyethylene bags. As previously discussed (Section 4.2.1.1), water contact with these targets will not be great enough for leaching to occur, especially since the double glass/polyethylene barriers can further reduce the likelihood of any possible water contact. The gaseous release of tritium will be the pertinent release and two cases will be considered: (1) the caps of the jars provide no barrier to gaseous release, and (2) the caps contain a polyvinyl chloride (PVC) film which acts as a permeability barrier to the tritiated species in the jar.

The targets consisted of copper tubing backings with titanium powder deposits which had been reacted with tritium, i.e., the tritium analog of titanium hydride had been formed. These targets, in practice, were used in vacuum systems; they were not exposed to air because oxidation of the titanium occurs (TiO_2 is thermodynamically much more stable than the hydride).[25] Upon disposal of these targets, exposure to air occurs. The outer layer of tritiated titanium would be expected to convert to TiO_2 to give an oxide coating. Thus, there might have been an initial loss of tritium from the target before it was ever packaged due to the oxidation reaction which displaces tritium on the titanium surface. Further loss of the tritium might occur upon oxidation of the inner layers of tritiated titanium but such a process would be limited by the permeability of the oxide layer to oxygen. Given the corrosion inhibition that generally results from the presence of an oxide layer on metals, it is likely that further oxidation would not occur as rapidly.

Loss of tritium from these targets can be estimated by extrapolation of data on tritium loss from targets after exposure to air as a function of time.[26] The percent of initial tritium remaining on a target at the end of one year is estimated to be 58%. The activity is lost in two ways: a 5% loss occurs in the first five days and is then followed by 0.1% loss per day thereafter. It is probable that some of the targets would have been exposed to air for more than five days before burial, in which case production of H-3 would have been at the lower rate of 0.1% per day from time of burial. However, since the number of targets, if any, in this category is not known, the conservative position is taken that all targets had the H-3 production rate of 5% in five days followed by 0.1% per day after they were buried.

Assuming the jar caps provide no barrier, the tritium activity released from the targets should all escape the jars and result in a release of 8.8 Ci the first year followed by 7.6 Ci the second year and 4.7 Ci the third year. Within three years, all of the activity will have escaped from these packages. If the jar caps contain PVC films, the tritium activity release will be limited by the permeability of the films. Local activity release from the targets will mainly be as T_2. There is sufficient water vapor in the

jars (assuming 370-cm^3 jar, 6-cm diameter cap, 5 Ci/jar) to allow exchange of the tritium with water to occur.[26] Two situations have been considered: (a) the exchange rate is fast, i.e., essentially all of the T_2 given off from the target becomes HTO, and (b) the exchange rate is slow, i.e., T_2 remains as T_2. In both situations, the exchange rate is compared to the rate of loss through the polyvinyl chloride film.

The permeabilities of polyvinyl chloride to H_2O[27] and H_2[28] are, in [cc(STP)$cm^{-2}yr^{-1}cm^{-1}$], 2.26×10^{-2} and 2.11×10^{-1}, respectively. It is assumed that these values change very little in going from H_2O to HTO and from H_2 to T_2. For fast exchange of T_2 with H_2O, (a), the tritium releases would be (in Ci): 3, 10, 4, 3, and 1 for the first through fifth years, respectively. In the fast exchange instance, all of the tritium would escape from the package within five years. For slow exchange, (b), the tritium releases would be just the amount produced, i.e., the polyvinyl chloride film permeability is high enough for T_2 that no effective barrier would exist in this case.

For the purposes of modeling the release, the more conservative scenario was adopted. Therefore, for slow exchange with water, the tritium releases would be in (Ci): 8.8, 7.6, and 4.7 for the first three years, respectively, so that essentially all the tritium would be released in three years. The incremental fraction releases then for Case 3 (Section 4.4.10) are 0.42, 0.36, and 0.22 for years 1, 2, and 3, respectively, and zero thereafter. Given the short release period, decay of the H-3 has been neglected.

4.4.1.2 Targets From Other Shippers

Six other generators besides BNL shipped targets to Sheffield -- see Table 4.5. Purdue University and University of Wisconsin each shipped $\geq$100 Ci. Primary containment of the Purdue targets was essentially the same as that for the BNL targets, but the secondary containers were metal paint cans inside carbon steel drums instead of concrete vaults. University of Wisconsin targets were shipped in metal paint cans, either in fiberboard boxes or with no secondary containment. Primary containment for the other shipments is not known, but they were all shipped in standard steel drums.

The Purdue shipments to Trench 2 are treated in the same manner as the BNL targets. This leads to annual releases, if fast exchange of T_2 with H_2O is assumed, of 14, 48, 19, 14, and 5 Ci for the first five years, and of 42, 36, and 22 Ci for the first three years for the case of slow exchange. All the other packages are considered to have had no effective barrier to release of gaseous tritium, so the release from all of them is considered to follow the experimentally-determined rate discussed in the previous section (4.4.1.1)[26] The estimated annual release rates are given in Table 4.6 for all targets, using the slow exchange case for BNL and Purdue targets.

Table 4.6

Estimated Annual Releases From Tritium Targets

Trench	Initial Target Activity (Ci)	Year	Total Annual Tritium Release From Targets (Ci) 1	2	3
1	120		50	44	26
2	120		50	44	26
7	13		5.3	4.7	3
11	14.2		5.8	5.1	3.3
14A	25.9		10.8	9.3	5.8
23	2		0.8	0.7	0.5

4.4.2 T_2O/HTO Sorbed on Desiccant

4.4.2.1 BNL van de Graaff Scrubbers

Two Drierite scrubbers used for removing T_2O/HTO from the vent line of BNL's van de Graaff accelerator were shipped to Trench 14A (see Table 4.5), one in a vault with 12-in. walls and one in a vault with 6-in. walls. As discussed in Sections 4.2.1.1 through 4.2.1.3, liquid release of H-3 from vaults can be disregarded, and gaseous release will not be affected by the wall thickness, i.e., neither 6-in. nor 12-in. walls will be a significant barrier to gaseous release. Drierite is a commercially available desiccant composed of anhydrous $CaSO_4$ and a moisture-indicating dye. Tritium gas (T_2) had been used as the source of tritons in the van de Graaf. The scrubber consisted of a cylindrical Lucite tube 3-in. O.D. with a 1/8-in. wall, and 5-3/8-in.-long, gasketed to aluminum end plates, each containing tabulation to a brass valve with metal seat. When a color change from blue to pink indicated useful life of the Drierite was nearing its end, the scrubber was simply valved off, removed from the van de Graaff equipment line, triply bagged in polyethylene, and sent for disposal. As with the concrete vault, the triple bagging does not inhibit escape of any gaseous tritium species released from the scrubber. Thus, the problem in estimating H-3 release from the whole package is to determine the release rate from the scrubber.

There are two main parts to this problem: determination of the rate at which tritiated gas or vapor species are formed, and of the rate at which they permeate through the 1/8-in. lucite scrubber wall.

(i) Rate of formation of gaseous T_2/HT by H-3 beta (β^-) radiolysis of T_2O/HTO.

As an approximation, it will be assumed that half of the tritium β-energy is deposited in the water species absorbed on the $CaSO_4$. The

scrubbers were replaced when half the contents, approximately 260 g, had changed color. The amount of water absorbed on the $CaSO_4$ has been estimated, by observation of the amount required to give a distinctly pink color, to be between 3 and 5%. The conservative position, since it leads to a higher production of T_2/HT gas, is to take the smaller value. This leads to 7.8 g or 0.43 moles of H_2O absorbed by $CaSO_4$ in the scrubber. The tritium content of the first scrubber is taken as 34 Ci averaged over the first year (35 Ci initially), which is equivalent to 0.6 mmol T_2O. Thus, the mole fraction of tritiated water species can be taken as 1.4×10^{-3}. For estimating the radiolysis yield of tritiated gas species (T_2/HT), it is not necessary to know the proportions of T_2O and HTO, or of T_2 and HT, since we estimate the release rate in terms of curies.

Total energy deposition in aqueous species from 34 Ci of H-3 in a year, assuming each β-particle has the average energy of 5.7 keV, is 1.1×10^{23} eV. Taking $G_{H_2} = 0.53$,[29] the production of hydrogen gas in the first year would be 0.94 mmol. On the basis of statistics, the mole fraction of tritiated hydrogen gas species will be the same as that for tritiated water species, or 1.4×10^{-3}. Therefore, the tritium production rate would be 1.3 μmole/yr, which is equivalent, for T_2, to 75 mCi/yr.

(ii) Rate of gaseous T_2/HT escape from the scrubber.

The rate of gas permeation through the aluminum end plates and brass valve seats is negligible in comparison to permeation through the Lucite walls of the scrubber. The rate of permeation of gas through plastic is given by the equation:

$$\frac{V}{t} = \frac{PAH}{\ell} \qquad (4.17)$$

where

V is the volume of gas permeating through the plastic.

t is the time in seconds.

P is the permeability constant in $cc(STP)cm \cdot cm^{-2}s^{-1}atm^{-1}$.

A is the area of the plastic in cm^2.

ℓ is the thickness of the plastic in cm.

H is the pressure head in atmospheres.

No value of P has been found for permeation of hydrogen through lucite, but values for other polymers are in the range of 10^{-7} to 10^{-9} in the units given above.[28] As a conservative approximation, 10^{-7} will be assumed. The value for time is taken as 1 yr (3.1×10^7 s), and the values of the other parameters are

$A = 300\ cm^2$

$\ell = 0.32\ cm$

$H = 5.5 \times 10^{-5}$ atm.

H is an approximation, which is simply the partial pressure exerted by half the gaseous tritiated hydrogen, based on T_2 produced in a year in the 300-mL void volume of the scrubber. From these values, the volume passing through the lucite in a year corresponds to approximately 0.16 mL of T_2, or approximately 400 mCi/yr.

This amount of tritiated gas exceeds the total amount estimated to be produced by β-particle radiolysis in a year from either scrubber - 75 mCi for one and 165 for the other. The calculation indicates that at the given pressure head, more gas can permeate the Lucite than would be available. Thus, the gas responsible for the pressure head must be lost through the Lucite almost as quickly as it is produced; no such pressure head can actually develop. Given the rapid transport of tritium through the Lucite cylinder, failure of or leakage through the gaskets at the aluminum end plates does not need to be considered. Effectively, all of the T_2 and HT produced by radiolysis can escape the scrubber by permeation through Lucite at a rate consistent with its generation rate. Thus, an initial release of inventory is estimated at approximately 75 mCi/yr and 165 mCi/yr, respectively, for the two scrubbers.

4.4.2.2 Sorbed T_2O/HTO Shipped by University of Wisconsin

The exact specifications of the scrubbers shipped by the University of Wisconsin are not known, so for the sake of simplicity, it is assumed that they were nearly enough like those used at BNL to be treated in the same way. Secondary containment used by the two generators (see Table 4.5) differed greatly, but neither type presented a significant barrier to gaseous release. Thus, the incremental fraction release of T_2/HT from the University of Wisconsin packages is assumed to be the same as that from the BNL scrubbers.

In the case of the University of Wisconsin packages, however, the scrubbers were susceptible to contact with water from a relatively short time after burial. Fiberboard boxes are assumed to be breached by the first contact with water, and the threaded end caps of the iron pipes would provide little, if any, barrier to liquid water. Thus, the only barrier to water would be the scrubbers themselves. The plastic bodies and metal end plates and valves would be expected to last many years, but the gaskets sealing the end plates to the bodies could well have a relatively short lifetime. The gasket material would most likely be rubber, but the particular material is not known. The gaskets in the trench environment would not be subjected to the heat and sunlight of above-ground conditions, but they would be exposed to radiation from neighboring packages. Once they failed, the entire contents of the scrubbers can be considered to be released as HTO to the trench water in the first year after their failure.

Thus, the most probable scenario for the release of tritium consists of relatively small amounts of gaseous release for several years, followed by a large liquid release of all the remaining tritium in one year. The exact time at which this liquid release would occur, due to failure of the rubber gaskets, is subject to so much uncertainty that no reasonable estimate can be made. There is also the possibility of mechanical or other failure (e.g., during shipping or emplacement), which would result in total release in the first year. The impact of the failure time chosen on the overall release for Trench 2 is discussed in Section 4.4.10.

4.4.3 Luminous Paint

This category of tritium waste contributed the largest curie amount of all the categories we have identified, approximately 850 Ci out of the total 2350 Ci shipped to the eight trenches covered by our study. Of this amount, Luminous Processes, Inc. shipped approximately 750 Ci. The shipments of Luminous Processes and of the other two generators, Timex Corporation and Illinois Institute of Technology, are listed in Table 4.5.

Until July 1976, Luminous Processes, Inc. shipped its waste in 4.5 ft^3 fiberboard boxes, while after that date, it used standard 55-gal drums. That is, shipments to Trenches 1, 2, 24, and 25C were made in boxes, and shipments to Trenches 23 and 14A were made in drums. The shipment from Timex Corporation was a single drum. It is uncertain whether the luminous paint in the single shipment sent by the Illinois Institute of Technology was contained in one or several 55-gal drums or was contained in a metal cask. However, for the purpose of this study, the conservative assumption will be made that this paint was buried in one 55-gal drum. The paint in the waste from Timex and Luminous Processes was mostly in the form of thin films of dried paint, but the Illinois Institute of Technology shipment consisted of unused bulk paint. In the treatment which follows, possible differences in H-3 release rates between these two cases are considered.

Since over 400 Ci of Luminous Processes waste was buried in fiberboard boxes, which are assumed to be accessible to water on emplacement, leaching must be considered as a potential release mechanism. From all the information we have been able to assess, it appears that H-3 release from leaching of luminous paint will be small in comparison with the amount escaping by gaseous release. For one thing, government regulations require that luminous paints or compositions must not leach or exchange tritium when immersed in water. The words "to a significant extent" should be added, since obviously some leaching and/or exchange will occur in time. However, apparently the rate is very low. This is supported by H-3 leaching studies done with tritiated polyacetylene.[30] The leach rate was measurable but so low that it was stated that the tritiated polyacetylene was "essentially non-leachable." The tritiated polymers used in luminous paints would be expected to behave similarly with respect to tritium loss, thus leaching is not considered further.

Information from Timex (Letter from Daniel H. Woods to D. R. MacKenzie, April 21, 1983, reproduced in Appendix B) detailed the process used by Timex,

from the mixing of the paint to the disposal of the H-3 waste. It is stated that, "the tritium waste consisted of luminous paint on scrap hands and dials, paper tissue used in cleaning, gloves, shoe covers, and glass shipping vials." The form of the waste in the Luminous Processes shipment to Trench 1 was described on the RSR as "luminous scrap dials and wipe tissue," and all their shipments are assumed to have been the same. Thus, the waste paint from these two generators can be taken to have been mostly in the form of relatively thin films, on discarded dials and other equipment, brushes, paper, rubber gloves, paint containers and lids, and other objects.

Gaseous tritium release from paint in the form of thin films has been determined experimentally. Table 4.7 contains the results of a study carried out on typical paints from four countries.[31] Note that the ratio between the rate of tritium loss from the paint and the tritium content of the paint (H-3 loss/H-3 content) is independent of the tritium content, the specific activity of tritium in the paint, and the painted area. These observations indicate that the rate of tritium loss is first order in tritium and that the mechanism by which it is lost does not involve radiolysis. Accordingly, a first-order rate constant (k_g) may be determined for the release of gaseous tritium from dry tritiated paint by using the equation

$$\text{Average (H-3 loss/H-3 content)} = k_g\ (1.0 \times 10^{-3}\ \text{Ci}) \qquad (4.18)$$

which results in a value of $4.5 \times 10^{-2}\ \text{yr}^{-1}$ for k_g. The half-life for gaseous tritium release determined from this rate constant is 15 yrs which is right in the middle of the range for this parameter (i.e., between 12 and 18 yrs) quoted by the Health Physics consultant to the Timex Corporation (see Letter from Daniel H. Woods to D. R. MacKenzie, April 21, 1983, reproduced in Appendix B).

Table 4.7

Gaseous Tritium Loss Rates From Surfaces Painted With Tritiated Paint[a](31)

Source of Paint	H-3 Content (mCi)	Specific Activity (mCi/g)	Painted Area (cm^2)	H-3 Loss (μCi/day)	H-3 Loss/H-3 Content (μCi/mCi-day)
Japan	24	1250	1.7	3.1	0.13
U.S.A.	25	100	6.9	3.1	0.12
Germany	25	250	2.1	3.2	0.13
England	53	250	4.2	6.0	0.11
Average	----	----	----	----	0.12

[a]Presumably measured after the paint had dried.

Accordingly, if gaseous release is the only mechanism by which tritium can be lost from this waste, then the amount of tritium left in the waste at a time (t) after it has been buried [I(t)] is determined by

$$I(t) = I_o \exp[-(k_g + \lambda)t] \tag{4.19}$$

where I_o is the amount of tritium which was present in the waste when it was buried and λ is the radioactive decay rate constant of tritium. The rate of tritium release (R_g), in this case, is given by

$$R_g = k_g I(t) \tag{4.20}$$

which may be integrated to give the amount of tritium released (AR_g) up to t

$$AR_g = \frac{k_g}{\lambda + k_g} I_o[1 - \exp(-(\lambda + k_g)t)] \tag{4.21}$$

Since the cumulative fraction release of tritium by this mechanism (CFR_g) is defined by

$$CFR_g = \frac{AR_g}{I_o} \tag{4.22}$$

it may be determined by

$$CFR_g = \frac{k_g}{\lambda + k_g} [1 - \exp(-(\lambda + k_g)t)] \tag{4.23}$$

Table 4.8 contains values of CFR_g calculated by use of Equation 4.23 for several times after waste burial. No credit has been given to drums for containment of gaseous release. Note that, because of radioactive decay, CFR_g changes hardly at all after 50 yrs.

The luminous paint sent by the Illinois Institute of Technology was unused (John F. Smalley, Memo to File, April 1, 1983, reproduced in Appendix B), i.e., it was still in solution. Since hydrogen gas is not very soluble, it is concluded that, on the time scales which are presently of interest, the paint's (organic) solvent presents no barrier to tritium gas (either HT or T_2) release. Thus, it will be assumed that the rate of release of tritiated gas from unused luminous paint is the same as that determined above for surfaces covered with dried paint. (If, however, the tritiated gas produced is not HT or T_2, then the solvent may hold up the tritium release. The present assumption is, in that case, conservative.)

Table 4.8

Cumulative Fraction Release (CFR_g) of Tritium From Luminous Paint

t(yr)[a]	CFR_g[b]
1.0	4.3×10^{-2}
5.0	1.8×10^{-1}
12.3[c]	3.2×10^{-1}
24.6	4.1×10^{-1}
50.0	4.4×10^{-1}
100.0	4.5×10^{-1}

[a]Time after waste burial.
[b]Calculated using Eq. (4.23), with $k_g = 4.5 \times 10^{-2}$ yr^{-1} and $\lambda = 5.6 \times 10^{-2}$ yr^{-1}.
[c]Tritium's radioactive decay half-life.

4.4.4 HTO in Concrete

BNL disposed of two packages of H-3 waste listed in Table 4.5 as HTO in concrete. Both packages were prepared in the same way. In each case, the H-3 was in the form of T_2O in a small glass bulb. The bulb was broken under freshly poured concrete in a 5-gal paint can, the concrete allowed to harden, and the lid fastened. The paint cans were shipped in BNL concrete vaults with 6-in. walls.

Release from the paint cans must be considered to be in two stages, one before and one after burial. Five months passed before the waste in the 67 Ci package was buried. During that five-month period, it is possible that substantial tritium may have been released from the package. This situation would have resulted from evaporative pore water loss through a faulty seal of the 5-gal can. The cans that were used are of a fairly common type. The lids have elastomer gaskets, and the lids are held in place by numerous small metal tabs. Commercial machine closure of this type container provides adequate sealing to prevent the loss of volatile components in the short term (several years). The quality of the seal produced by hand closure, which was the case for the BNL waste containers, is uncertain. Two polar cases are, therefore, considered in the short term: (1) Hand closure provided an adequate container seal and the inventory was contained, and (2) hand closure resulted in a faulty seal and no containment of evaporative water loss was provided by the container. A discussion of the latter is provided below.

Complete exchange of the H-3 with the water in the cement will be assumed as the conservative approach since it leads to a lesser amount escaping

before burial, or a larger amount buried. In practice, only free pore water evaporates. Some of the water used to mix the cement will have bonded to the cement before mixing of the T_2O in the cement occurred, so that a larger fraction of the H-3 would probably be with the pore water than would be calculated assuming complete exchange with the total water. Then as pore water evaporates taking its H-3 content with it, less H-3 will be available for exchange with bound water so complete exchange would have never been achieved.

Assuming a water:cement ratio of 0.6 for the concrete in the paint can, the fraction of water available to evaporate is 0.25 (i.e., the water/cement ratio needed for complete hydration is assumed to be 0.35).[14] In five months, it was assumed that essentially all this water will have evaporated and carried with it a fraction equal to 0.25/0.60 = 0.42 of the initial H-3, assuming the complete exchange discussed in the previous paragraph. Thus, after five months, only 58% of the original tritium will remain in the waste container.

After loss of pore water, which should dominate short-term releases, the residual tritium releases need to be addressed. The release of tritiated vapor from a cylindrical cement block has been studied by Emelity and coworkers.[32] The cement in this block had a water/cement ratio of 0.3 (which means that all of the water is probably hydration water - i.e., no tritium will escape by evaporation) and had been cured for an unspecified period of time. The cumulative fraction release (CFR) data from this study is fit (for times greater than five days after the removal of the cement block from its container) by

$$CFR = (S/V)D't^{1/2} \qquad (4.24)$$

where S/V is the ratio of the cylinder's exterior surface area to its volume and D' is a property of the waste form solidification agent (i.e., cement). The data in Reference 32 give $(S/V)D' = 0.74\%/day^{1/2}$.

It is assumed that these results may be applied to the release of tritiated vapor from the BNL cement waste forms and that the metal paint cans holding the BNL waste forms do not impede the release of the tritiated vapor to the interior of the vault. Thus, since S/V for the cylinder in Reference 32 is $\sim 1.2\ cm^{-1}$* and S/V for the BNL waste forms is $\sim 0.21\ cm^{-1}$, the release of tritiated vapor from the BNL waste forms may be described by

$$CFR = (2.5\%/yr^{1/2})t^{1/2} \qquad (4.25)$$

The amount released (AR) from the cement waste forms contained in this vault, therefore, is

$$AR = (1.0\ Ci/yr^{1/2})t^{1/2} \qquad (4.26)$$

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*Telephone conversation between Ruth Kempf (BNL) and L. A. Emelity (Los Alamos Scientific Laboratory) on March 31, 1983.

Equation 4.26 has been corrected for the fact that only 58% of the original amount of tritium is left in the paint can when it is buried. Thus, in the first through fourth years, the releases from the 67 Ci package will be 1.0, 0.41, 0.32, and 0.27 Ci, respectively. The release rate is described by the derivative of Equation 4.26. It should be noted that the form of Equation 4.24 is identical to the short-term solution for plane sheet diffusion of a substance from a membrane - a model which is often used to describe radionuclide leaching from solidified waste forms.[33] Hence, it seems reasonable to expect this relationship to be valid only for the initial loss of approximately 20% of the bound tritium activity. Beyond 20% release, the method of release prediction proposed in the draft standard on low-level waste leaching, ANS 16.1,[34] appears adequate.

Production of HT/T_2 gas by β^- radiolysis of the tritiated water in the concrete waste form of the 67 Ci package can be estimated in a manner similar to that used in Section 4.2.2. The fraction of energy absorbed by tritiated species has been taken as the weight fraction of tritiated species to total concrete weight. The density of hydrated concrete having no pore water is approximately 1.52 g/cm^3.[18] The volume of the concrete form was 5 gallons and contained 39.1 Ci of tritiated water. The fraction of β^- energy absorbed by tritiated species will then be 8×10^{-7}. Hence, the amount of HT/T_2 gas produced in the first year is 7×10^{-5} Ci or 0.07 mCi. Compared to the 1.0 Ci first year expected tritium release (due to a combination of mechanisms) derived from the experimental data of Emelity,[32] this release from radiolysis of the hydration water is insignificant.

In summary, it is not possible to predict the actual tritium releases from these 5-gal cement-filled containers with any great certainty due to the uncertainty in the quality of container seal. Therefore, two limiting cases were considered. In both cases, vapor phase transport is dominant, radiolysis is negligible, and the concrete vault ineffective as a barrier.

In the first case, lid seals are assumed to be adequate for short-term storage and shipping. Thus, the total tritium activity recorded on the RSRs is assumed to have reached and been emplaced at the Sheffield site. Container failure through seals and/or corrosion is assumed to occur shortly after emplacement. This leads to approximately 40% tritium loss through evaporation of pore water occurring rapidly. This is believed to occur in the first year after container failure and is based upon the permeability considerations in Section 4.2.1.2 and the anticipated site environment. Superimposed upon the evaporative loss is the release of bound tritium which is predicted by Equation 4.26.

In the second case, no reliance on lid seals, the evaporative loss is assumed. Therefore, during short-term storage and shipping of these BNL waste packages, tritium loss occurs. Approximately 40% of the inventory of the cement waste package is <u>not</u> emplaced at the Sheffield site, and is therefore omitted as a source term to the disposal trench. The release of remaining tritium (40 Ci) from this package, approximately 60% of the RSR inventory, is estimated by the application of Equation 4.26. In all likelihood, the actual

and site release are intermediate between the bounds given in the two cases above. However, it is not currently possible to realistically describe the actual condition of these packages. Taking the conservative approach, calculation of annual releases to the trenches in a subsequent section (4.4.10) makes the assumption that the lid seals hold only until the waste is buried.

4.4.5 Generalized Laboratory Trash

Table 4.5 lists a large number of shipments with packages of relatively large curie amounts which contained either waste of a general nature, such as glassware, paper, plastic, animal carcasses, etc., contaminated with labelled chemicals, or waste about which insufficient information could be obtained to place it in one of the special categories. As indicated in the footnotes to the table, there were a number of shipments >1 Ci, but with lesser amounts than those listed, which are included in our treatment.

The 4.9 Ci BNL shipment constitutes a special case and will be considered first. It is assumed that no H-3 releases will occur by leaching in the time frame of interest, and the only release mechanism operating is escape of tritiated gases out of the plastic bag and thence, out of the concrete vault. Formation of tritiated gas by radiolysis can be estimated in the same way as formation of T_2/HT from T_2O/HTO on the drierite surface in the scrubber considered in Section 4.4.2.1, only in this case all the gas formed will escape in a shorter time. This leads to a value for tritium-containing gases formed, and escaping, of 1.9×10^{-8} Ci/yr.

Tritium releases that appear to follow first order kinetics with a rate constant approximately the same as that of natural radioactive decay have been observed with tritiated polymers in air.[31] This could be a result of a process in which hydrogen-containing species in the air come in contact with the polymer surface, undergo exchange with the tritium, and then are removed by diffusion. This is discussed in Section 4.4.3. Such a process should occur in any situation in which tritiated organic species are in contact with hydrogen-containing species in air. The contaminated lab trash (tritiated compounds on cellulose and glass) can be considered analogous to the tritiated polymers. Thus, this package would be expected to release tritium in a similar manner. We have taken as the first order rate constant for tritium loss:

$$k = 4.5 \times 10^{-2}\ yr^{-1}. \qquad (4.27)$$

The tritium release due to this process is 0.2 Ci in the first year. Compared to this value, the release of tritium produced by radiolysis is insignificant.

It has been assumed that the remaining tritium-containing waste, could all be approximated as generalized lab trash. The part of the waste in fiberboard boxes comes under Case 8, while that in drums is treated in the same way as the liquid wastes solidified at Sheffield, which are covered by Case 2. This means that 15% of the drums are assumed to be breached on emplacement and their contents immediately leached. Thus, the incremental fraction release for the first year includes 15% of the waste plus the incremental release from

the other 85% of the waste. The expression for IFR is then the same as that given by Equations 4.15 and 4.16 in Section 4.3.2.

4.4.6 Oil Sorbed on Vermiculite

The waste shipped by the University of Wisconsin differed somewhat from that of Caterpillar Tractor Co. (see Table 4.5). The former consisted of contaminated pump oil, while the latter was actually tritiated oil.

The trititated oil is assumed to be composed of hydrocarbons and can be assumed to behave in a manner similar to the unused bulk luminous paint discussed in Section 4.4.3. This position is based on the high probability that the relatively high molecular weight hydrocarbons would act like the tritiated polymer molecules in luminous paint with respect to tritium release. In that case, H-3 release by leaching would be insignificant (Section 4.4.3) and annual incremental fraction releases would be the same as those for the luminous paint.

The contaminated pump oil is treated as generalized laboratory trash since the contaminants are unknown. Some of them may be organic compounds of a type which should be treated like luminous paint. Also, undoubtedly some of the pump oil would have become tritiated. However, since the exact nature of this waste is not known, the conservative position requires that we treat all of it as general laboratory waste (see Section 4.4.5) even though part of it may release H-3 at a lower rate.

4.4.7 T_2/HT Gas in Steel Cylinders

A shipment by BNL to Trench 14A consisted of 129 Ci of T_2/HT gas contained in two Hoke isotope containers in a standard 5-ft x 5-ft x 6-ft BNL concrete vault. The two containers each consisted of an inner stainless steel cylinder jacketed within a stainless steel case. All welded construction was used throughout, and the inner steel cylinders were valved with special stainless steel valves. The leak rate of the system was checked by the manufacturer with a mass spectrometer using He gas, only those containers being passed from which no helium escape could be detected, with a detection limit of 10 microns $Hg \cdot ft^3/h$. Since the BNL containers had passed this test, it is considered that their leak rates were less than this value, which corresponds to 2.4×10^{-2} cc atm/yr of helium and less of other gases. Using this value as the leak rate for T_2 gas leads to an annual leak rate from each container of 1.2×10^{-3} cc/yr for the first year, since the T_2 partial pressure was approximately 0.05 atm. This represents a total volume of 2.4×10^{-3} cc/yr or 6 mCi in the first year, and correspondingly less in future years. Such amounts are too small to affect the total H-3 release to Trench 14A and are thus omitted from further consideration.

4.4.8 T_2/HT Gas in Glass Bulbs

Shipments of T_2/HT gas in glass bulbs were sent by Abbott Laboratories, Michigan State University and Univeristy of Illinois (Table 4.5). All the shipments can be treated in the same general way, and that of the

University of Illinois is used as an example. It consisted of five sealed glass bulbs containing 1 Ci each of tritium gas. Each bulb was wrapped in packing material and placed in a 2-in. diam piece of iron pipe capped off at both ends. The pipes were set on the bottom of a standard steel drum, and general laboratory trash piled on top.

In terms of H-3 release from the package, there are two polar cases: (1) all the bulbs could have remained intact during package emplacement in the trench, and (2) all the bulbs could have broken during emplacement. In the first case, rate of release would be determined by the rate of T_2 permeation through the walls of the glass bulbs. In the second case, the threaded end caps of the iron pipe would allow immediate release, and the only potential barrier to escape of H-3 to the trench would be the drum gasket.

1st Case - All Bulbs Remain Intact

The rate of permeation of gas through glass is given by the equation:

$$\frac{V}{t} = \frac{PAH}{\ell} \qquad (4.28)$$

where

V is the volume of gas permeating through the glass in cc.

t is the time in seconds.

P is the permeability constant in $cc(STP)cm \cdot cm^{-2}s^{-1}atm^{-1}$.

A is the area of the glass in cm^2.

ℓ is the thickness of the glass in cm.

H is the pressure head in atmospheres.

The value of P for permeation of D_2 through borosilicate glass is 10^{-13} in the units given above.[28] The same value is assumed for T_2. The value for time is taken as 1 yr, $t = 3.1 \times 10^7$ s, and the values of the other parameters as

$A = 70\ cm^2$.

$\ell = 0.15$ cm.

$H = 0.02$ atm.

These values are arrived at on the assumption that the glass bulbs inside the 2-in. diam iron pipes were approximately 1-in. diam and 3-in. long, with a 1.5-mm wall thickness. The permeation rate will not be particularly sensitive to the size of the bulb, since, as the value of A increases, the volume increases and the value of H decreases to compensate.

Using the values above, the permeation rate is estimated to be 29 nL/yr or approximately 8 ng/yr. This is equivalent to approximately 0.08 mCi/yr for each bulb, or 0.4 mCi/yr for all five. Thus, the maximum amount of H-3 which could escape in Case 1 is <1 part in 10^4 per yr.

2nd Case - All Bulbs Break During Emplacement

No strong argument can be made that the drum gasket would provide a serious barrier to escape of tritium gas. In fact, the opposite could be argued more effectively, since DOT regulations (49 CFR 178-118-66) require that a hydrostatic pressure of 15 psi be sustained for only 5 minutes, and in practice, even this criterion is seldom met. For example, in tests done on 30 standard drums (ten each from three different manufacturing lots), only one drum passed the test.(35)

Thus, the conservative position requires that, in the second case, all the H-3 will escape from the drum within the first year. An appreciable fraction of the release could even have occurred before the drum was backfilled in the trench, depending on the leak rate and the length of time before backfilling. There is, however, no way of determining this, so the conservative approach from the point of view of groundwater uptake is to assume release of all 5 Ci to the trench soil in the first year.

It is concluded that the two extreme cases (all bulbs break or all remain intact) lead, respectively, to release rates of the whole inventory in the first year, or <1 part in 10^4 per year indefinitely. The actual situation is probably somewhere between these two extremes. This does not hold, of course, for packages containing only one bulb, which was the case for the other two shippers. Here the two polar cases apply. In the case of the Abbott packages, the caps were apparently welded to the pipes by NECO after reaching Sheffield (see Letter From R. L. Frederickson to D. R. MacKenzie, August 23, 1983, reproduced in Appendix B). If the welds were sound, essentially no release would occur until the pipe material was breached by corrosion, at which time either slow release would begin, if the glass bulbs were intact, or all the gas would be released, if they had broken. For simplicity, however, we have chosen to treat the Abbott case in the same way as the others for purposes of total release to Trench 1, so all these shipments are treated as belonging under Case 8 in the list of Cases given in Section 4.4.

4.4.9 Liquid Waste

The large shipments of liquid waste which were solidified by NECO include both those listed in Table 4.5 and a number containing lesser amounts (but still $\geq$1 Ci). These fall under Case 2 and are treated in the same way as the liquid waste from shipments <1 Ci, which was discussed in Section 4.3.2. Besides these shipments, one large shipment of tritiated water which was not solidified must be considered.

This shipment by Mallinckrodt Nuclear to Trench 7 had a H-3 activity of 9.6 Ci as HTO. The water was contained in 10-mL glass vials with rubber stoppers and metal crimp caps (see Memo to File, of April 11, 1983, reproduced in Appendix B). It is expected that this method of closure would provide some inhibition of tritium release, but in order to determine the degree of inhibition, the container integrity must be assessed more thoroughly than was possible. It has, therefore, been conservatively assumed that the total vial contents were released to the packing material. The waste then resembles the generalized contaminated laboratory trash and, since all of these Mallinckrodt wastes were packed in fiberboard containers, this results in total loss of activity in the first year.

4.4.10 H-3 Releases by Case and by Trench

In preceding sections, expressions have been developed for the IFR for each of the eight cases to which the different types of waste have been assigned. These cases, and the equations for IFR for each are listed below. In all cases, $\lambda = 5.6 \times 10^{-2}\ yr^{-1}$.

Case 1, Shipments <1 Ci, Solid

$$IFR(t) = 0.49 + 0.51 \times \text{Drum Release}(t) \qquad t = 1$$
$$IFR(t) = 0.51 \times \text{Drum Release}(t) \qquad t > 1$$

Case 2, Liquid Shipments <1 Ci and >1 Ci, General Waste in Shipments >1 Ci

$$IFR(t) = 0.15 + 0.85 \times \text{Drum Release}(t) \qquad t = 1$$
$$= 0.85 \times \text{Drum Release}(t) \qquad t > 1$$

Case 3, Targets

$$IFR(1) = 8.8/21.1$$
$$IFR(2) = 7.6/21.1$$
$$IFR(3) = 4.7/21.1$$
$$IFR(t > 3) = 0$$

Case 4, Scrubbers

$$IFR(t) = 0.039\ (\exp(-\lambda(t - 1)) - \exp(-\lambda t))$$

Case 5, Tritiated Organics

$$IFR(1) = \frac{k}{\lambda + k}\ (1 - \exp(-(\lambda + k)t)) \qquad t = 1$$

$$IFR(t) = \frac{k}{\lambda + k}\ (1 - \exp(-(\lambda + k)t)) - CFR(t - 1)$$

where $k = 0.045$

Case 6, Tritiated Cement

$$IFR(t) = 0.145(t^{1/2} - (t - 1)^{1/2})\ e^{-\lambda t}$$

Case 7, BNL General Waste, Shipments >1 Ci

$$IFR(t) = \frac{0.045}{\lambda + 0.045}\ (\exp(-(\lambda + 0.045)(t - 1) - \exp\ (-(\lambda + 0.045)t))$$

Case 8, Release During First Year (Broken Glass Bulbs, etc.)

$$IFR(1) = 1.0$$
$$IFR(t > 1) = 0$$

The "Drum Release" referred to in Cases 1 and 2 is the function derived in Appendix C for IFR_T. For these calculations, values of $IFR_T(t)$ were taken directly from Table C.1.

A FORTRAN program for the CDC 6600 has been written to give annual IFRs and CFRs for the eight cases, and to calculate annual incremental and cumulative releases for each trench. In order to obtain the incremental and cumulative releases for a specific trench, the IFRs and CFRs for each case have only to be multiplied by the amounts of waste assigned to the respective cases for that trench, and the results summed.

The program is reproduced as Table 4.9. The curie amounts assigned to the different cases for each trench are given in Table 4.10. The annual IFRs and CFRs for each case, and the annual incremental and cumulative releases for each trench have been calculated for a period of 90 years. The results are given on microfiche in Appendix D, Tables D.1 and D.2. An abbreviated form of Table D.2 (annual releases by trench) is given in Section 4.10, along with similar information for other isotopes of concern.

Releases to Trench 2 in the first few years are problematic because of the uncertainty connected with the fate of the University of Wisconsin gas scrubbers (Section 4.4.2.2). The most probable scenario is that release of gas according to the model developed for BNL scrubbers (Case 4) would occur for a period of several years until the gaskets fail, and then all the tritium would be relesed in the next year as HTO in liquid water. This assumption of complete release in one year is based on the fact that the fiberboard box containers would be expected to provide no barrier to liquid water. The time to failure of the gaskets is highly uncertain, and might even be ≤ 1 year, which would mean complete release in the first year. Since we can make no good estimate of the time, we have considered two extremes: (1) slow release according to Case 4 for 90 years, and (2) total release in the first year. The releases for Trench 2 given in Table D.2 use the first assumption. Both cases are given in the abbreviated table of tritium releases (Table 4.21) in Section 4.10. The actual situation presumably will be somewhere between the two extremes, with release of all the remaining H-3 occurring after some time probably no longer than 20 years, and most likely in a considerably shorter time.

Table 4.9

FORTRAN Code to Calculate IFRs and CFRs for Eight Cases, and Annual Incremental and Cumulative Releases for Each Trench

```
      PROGRAM TRIT(INPUT,OUTPUT,TAPE5,TAPE6,TAPE7)
      DIMENSION DR(90),TI(8,8),CFR(8,90),TN(8),AR(8,90),
     1TR(8,90),CI(8,8)
      REAL LAM,KG,K6,K7,IFR(8,90)
      LAM=0.057
      DO 50 I=1,90
      READ(5,20) DR(I)
   20 FORMAT(G7.1)
   50 CONTINUE
      DO 60 J=1,8
      DO 60 K=1,90
      IFR(J,K)=0.0
      CFR(J,K)=0.0
      AR(J,K)=0.0
      TR(J,K)=0.0
   60 CONTINUE
C     CALCULATE IFR FOR EACH CASE
   61 FORMAT(1X      "CASE ANALYZED")
      DO 70 M1=1,90
      IF(M1.EQ.1) IFR(1,1)=0.49+(0.51*DR(1))
      IF(M1.GT.1) IFR(1,M1)=0.51*DR(M1)
   70 CONTINUE
      WRITE 61
      DO 80 M2=1,90
      IF(M2.EQ.1) IFR(2,1)=0.15+0.85*DR(1)
      IF(M2.GT.1) IFR(2,M2)=0.85*DR(M2)
   80 CONTINUE
      WRITE 61
      IFR(3,1)= 8.8/21.1
      IFR(3,2)=7.6/21.1
      IFR(3,3)=4.7/21.1
      WRITE 61
      DO 90 M4=1,90
      M4M=M4-1
      IFR(4,M4)=0.039*(EXP(-1.0*LAM*M4M)-EXP(-1.0*LAM*M4))
   90 CONTINUE
      WRITE 61
      S5=0.0
      DO 100 M5=1,90
      KG=0.045
      CR=(KG/(LAM+KG))*(1.0-EXP(-(KG+LAM)*M5))
      IFR(5,M5)=CR-S5
      S5=S5+IFR(5,M5)
  100 CONTINUE
      WRITE 61
```

Table 4.9, Continued

FORTRAN Code to Calculate IFRs and CFRs for Eight Cases, and Annual Incremental and Cumulative Releases for Each Trench

```
      DO 110 M6=1,90
      K6=0.0145
      M6M=M6-1
      X1=(M6M**0.5)*K6
      X2=(M6**0.5)*K6
      IFR(6,M6)=(X2-X1)*(EXP(-1.0*LAM*M6))
  110 CONTINUE
      WRITE 61
      DO 120 M7=1,90
      K7=0.045
      M7M=M7-1
      AK7=LAM+K7
      IFR(7,M7)=(K7/AK7)*(EXP(-1.0*AK7*M7M)-EXP(-1.0*AK7*M7))
  120 CONTINUE
      WRITE 61
      IFR(8,1)=1.0
      WRITE 61
C      CALCULATE CFR
      DO 150 IS=1,8
      DO 150 JS=1,90
      JSM=JS-1
      IF(JS.EQ.1) CFR(IS,JS)=IFR(IS,JS)
      IF(JS.GT.1) CFR(IS,JS)=CFR(IS,JSM)+IFR(IS,JS)
  150 CONTINUE
      WRITE 151
  151 FORMAT(1X      'CFR'S CALCULATED')
C      CALCULATE TRENCH RELEASES
      DO 200 IT=1,8
      READ(5,160) TN(IT)
  160 FORMAT(A3)
      READ(5,170) (CI(IT,JT),JT=1,8)
  170 FORMAT(8G10.4)
      DO 180 LT=1,90
      DO 180 KT=1,8
      TR(IT,LT)=TR(IT,LT)+(CFR(KT,LT)*CI(IT,KT))
      AR(IT,LT)=AR(IT,LT)+(IFR(KT,LT)*CI(IT,KT))
  180 CONTINUE
      WRITE 181
  181 FORMAT(1X      'TRENCH RELEASES CALCULATED')
  200 CONTINUE
  210 FORMAT(1X,8X,3(9X,'CASE  ',I1,9X))
  220 FORMAT(1X,8X,2(9X,'CASE  ',I1,9X))
  230 FORMAT(1X,8X,3(7X,'TRENCH  ',A3,7X))
  240 FORMAT(1X,8X,2(7X,'TRENCH  ',A3,7X))
```

Table 4.9, Continued

FORTRAN Code to Calculate IFRs and CFRs for Eight Cases, and Annual Incremental and Cumulative Releases for Each Trench

```
  250 FORMAT(1X,1X,"TIME",3X
     13("INCREMENTAL",2X,"CUMULATIVE",2X),/,1X,"(YEARS)",
     22(2X,"RELEASE-H3"),2X,"RELEASE-H3",3X,"RELEASE-H3",2X,"RELEASE-H3"
     33X,"RELEASE-H3",/,1X,9X,1X,"(CURIES)",4X,"(CURIES)",3X,
     41X,"(CURIES)",5X,"(CURIES)",4X,"(CURIES)",5X,"(CURIES)",/)
  270 FORMAT(1X,2X,I3,2X,4(1P,4X,G7.1,1X),5X,G7.1,1X,5X,G7.1,1X)
  280 FORMAT(1X,1X,"TIME",3X
     12("INCREMENTAL",2X,"CUMULATIVE",2X),/,1X,"(YEARS)",
     22(2X,"RELEASE-H3"),2X,"RELEASE-H3",3X,"RELEASE-H3"
     3,/,1X,9X,1X,"(CURIES)",4X,"(CURIES)",3X,
     41X,"(CURIES)",5X,"(CURIES)",/)
  290 FORMAT(1X,2X,I3,2X,4(1P,4X,G7.1,1X))
C       OUTPUT RESULTS
      DO 400 LO=1,2
      LX=LO*3
      LY=LX-2
      WRITE(6,210) (NY,NY=LY,LX)
      WRITE(7,230) (TN(NX),NX=LY,LX)
      WRITE(6,250)
      WRITE(7,250)
      DO 400 LT=1,90
      WRITE(6,270) LT,(IFR(MY,LT),CFR(MY,LT),MY=LY,LX)
      WRITE(7,270) LT,(AR(MZ,LT),TR(MZ,LT),MZ=LY,LX)
  400 CONTINUE
      NV=7
      NW=8
      WRITE(6,220) NV,NW
      WRITE(7,240) TN(NV),TN(NW)
      WRITE(6,280)
      WRITE(7,280)
      DO 500 LW=1,90
      WRITE(6,290) LW,(IFR(NQ,LW),CFR(NQ,LW),NQ=7,8)
      WRITE(7,290) LW,(AR(NT,LW),TR(NT,LW),NT=7,8)
  500 CONTINUE
      STOP
      END
      FUNCTION IFA(N)
      IFA=1
      IF(N.EQ.0) GO TO 20
      DO 10 J=1,N
   10 IFA=IFA*J
   20 CONTINUE
      RETURN
      END
..
```

Table 4.10

H-3 Inventory for Each Trench for
the Eight Cases Requiring Separate Consideration

Trench	Case							
	1	2	3	4	5	6	7	8
1	2.8	24.1	120	0	136	0	0	27.4
2	7.1	232.7	120	193	29	0	0	47
7	2.7	10.3	13	0	0	0	0	.6
11	2.7	18.9	14.2	0	0	0	0	0
14A	7	51.7	25.9	112	225	162	4.9	5
23	4.3	28.9	2.5	0	152	0	0	0
24	3.5	55.6	0	0	256	0	0	0
25C	2.1	44.4	0	0	67.5	0	0	0

4.5 Estimated C-14 Releases

The C-14 wastes shipped to the trenches covered by this study are listed in Tables 2.13-2.15. Out of the estimated 104 Ci total activity inventory, Pathfinder Laboratories contributed just over half. This amount went to only three trenches - Trenches 14A, 23, and 24. One other trench (Trench 2) had a relatively large amount, approximately 28 Ci, composed mostly of shipments >1 Ci from Amersham/Searle and Mallinckrodt Nuclear. The remainder of the trenches held total amounts ranging from <1 Ci to about 4 Ci, mostly from shipments containing small amounts of general laboratory waste.

The wastes have been divided into two main categories: (1) solid organic wastes (Pathfinder, Amersham/Searle, etc.) and (2) miscellaneous C-14 contaminated wastes which included paper, glassware, animal carcasses, gloves, etc. The wastes which fall into the first category have been modeled to release their radionuclide activity by two mechanisms: (a) gaseous release from C-14 β^- radiolysis, and (b) release by contact with water through (1) simple loss-on-contact leaching and (2) waste volume removal "wash". Release from the miscellaneous or generalized wastes is based on a simple leaching model. This will be considered first.

4.5.1 Release From Low-Level General Waste

4.5.1.1 Estimation of Distribution of Package Types

To obtain an estimate of the percentage of fiberboard containers and 55-gal drums which contain C-14 wastes, a survey of the RSRs for Trench 25C was performed, using methods similar to those applied to H-3 Low-Level wastes and described in Section 4.3.1.1. Only those shipments which contained C-14 were included in the survey. It is assumed that shippers who disposed of C-14

wastes packaged their waste uniformly. The total solid volume per shipment was divided by the total solid packages to yield a volume per package. Shipments containing packages with average volumes above 6 cu ft and less than or equal to 9 cu ft were assumed to be comprised mainly of 55-gal drums. Shipments containing packages with average volumes of 3 to 5 cu ft were assumed to be comprised of fiberboard containers. The results of this survey are given in Table 4.11. The percentages of 55-gal drums and fiberboard containers are 80 and 20, respectively. By the same logic used in our treatment of the H-3 wastes, it is to be expected that all of the fiberboard containers and 15% of the 55-gal drums will release their contents during the first year. The remaining 85% of the drums will undergo corrosion and leaching and have been treated according to the model developed in Appendix C. The incremental releases from the generalized C-14 lab wastes are dependent on the C-14 activity distribution which is discussed in the next section.

Table 4.11

Distribution of Types of Packages
Containing C-14 in Trench 25C

Total Number of Solid Packages (either 55-gallon drums or fiberboard containers)	1000
Estimated Number of 55-Gallon Drums	800
Estimated Number of Fiberboard Containers	200
Fraction of Solid Packages which were 55-Gallon Drums	0.8
Fraction of Solid Packages which were Fiberboard Containers	0.2

4.5.1.2 Distribution of C-14 Activity by Trench

Since RSR listings rarely specified the particular isotope amounts in a package containing mixed isotopes, it is difficult to determine the exact C-14 activity in the widespread generalized wastes. To estimate this quantity, a survey of each trench inventory was performed in which C-14 activity entries on RSRs which did list C-14 and its amounts were totalled - except, of course, for shipments >1 Ci. For each trench, this value was then divided by the total activity (C-14 and the other radioisotopes present as well) of such shipments to give a ratio which is expected to reflect an approximate C-14 fraction for typical wastes. It was assumed that the RSRs which specifically listed C-14 were a random lot and that the C-14 distribution found from these

RSRs and represented by the ratio mentioned above was applicable to the entire contents of the trench. The trench activity totals for all shipments, which listed C-14 as a constituent, were then multiplied by this ratio to yield an approximate total C-14 activity in generalized lab waste for that trench. It is this amount which will be taken as spread throughout the trench in fiberboard containers (20% of the activity) and 55-gallon drums (80% of the activity).

4.5.1.3 Release From Generalized Laboratory Waste

The generalized waste is assumed to leach by release on contact with water. Releases of C-14 activity are expected in the first year to be mainly from fiberboard containers and the 15% of drums which were breached during emplacement in the trench. No releases will then occur until the fifth year when first penetration of the remaining 85% of the drums occurs. In subsequent years, release will steadily increase as the drums corrode with time. The treatment used is based on the model for activity loss from a carbon steel drum, developed in Appendix C. The expected C-14 activities calculated in Section 4.5.1.2 have been used in estimating the annual incremental releases and the results are given in Table 4.12. It can be seen from the table that the expected large first year release to all trenches occurs. This is followed by the 5-year induction period before corrosion penetrates the drums. Once pitting corrosion penetrates the drums, several years of very low release occur. This is because the first pitting areas are quite small, so very little water can enter the drum to cause release of activity. It should be noted that a fraction of the miscellaneous waste, which was shipped as liquid and solidified by NECO at Sheffield, would have been buried as concrete. The C-14 in the concrete would presumably be held back more than that in miscellaneous solid lab trash, so the assumption that the general waste consists of only solid contaminated trash is conservative.

Table 4.12

Incremental Annual Releases (in mCi) of C-14 From Low-Level Generalized Wastes

Incremental Fraction Release:		1.4×10^{-4}	1×10^{-3}	4×10^{-3}	7×10^{-3}	9×10^{-3}	1.2×10^{-2}	1.5×10^{-2}	1.7×10^{-2}	2×10^{-2}	2.3×10^{-2}
Year:	1[a]	6	10	20	30	40	50	60	70	80	90
Trench											
1	384 (240,144)	0(0.1)[b]	1	3	6	7	10	12	14	16	19
2	1760 (1100,660)	1	4	15	26	34	45	56	64	75	86
7	239 (149,90)	0(0.1)	1	2	3	5	6	7	9	10	12
11	1424 (890,534)	0(0.4)	4	12	20	28	36	44	52	60	68
14A	619 (387,232)	0(0.2)	2	5	9	12	16	19	23	26	30
23	1499 (937,562)	0(0.4)	4	13	21	30	38	47	55	64	72
24	800 (500,300)	0(0.2)	2	7	12	15	20	26	29	34	39
25C	160 (100,60)	0(0.05)	0(0.4)	1	1	3	4	5	6	7	8

[a]Numbers in ( ) represent releases from fiberboard containers and 55-gallon drums, respectively.
[b]The releases in ( ) are considered insignificant but are included for completeness.

4.5.2 Release From Shipments >1 Ci

The waste in these shipments is not, in general, well characterized. Information from the three largest generators is contained in a Memo to File by D. R. MacKenzie, September 1, 1983, reproduced in Appendix B. All the wastes are solid labelled organics, Pathfinder's being mainly aromatic. However, for none of the generators can estimation of both leaching release and gaseous release from the compounds be considered as straightforward releases, because the identities of the compounds, the specific amounts of each, and the distribution of these materials in any given waste package are all unknown. To be consistently conservative, we have taken the highest organic compound G-value for gas production for our radiolysis calculations.

The Pathfinder case will be used for the calculation of release due to radiolysis, and behavior of wastes from the other generators is assumed to be similar.

4.5.2.1 Release Due to β^- Radiolysis

The solid organics in Pathfinder waste were mainly aromatics, of specific activities ranging between 5 and 20 mCi per millimole of aromatic. An average specific activity of 12.5 mCi/mmol has been assumed for all of these wastes. The G-values for radiolysis for the C-14-containing organic compounds which may be present cannot be determined in the absence of a detailed description of the actual or even likely waste package contents. Thus, a G-value for the generation of simple carbon-containing gases (CO_2 and CH_4) will be assumed based on data for aromatic compounds available in the literature. Swallow[36] gives a range of G-values for total gas of between <0.012 and 0.26 molecules per 100 eV for aromatic hydrocarbons. The chief gaseous radiolysis product from these compounds is hydrogen and, where reported, the G value for CH_4 production may be more than a factor of two less. For benzoic acid, Swallow indicates that CO_2 is a major gaseous product and reports a G-value of 0.29 molecules per 100 eV for CO_2 production. In a more recent summary of radiation effects on materials, Kircher and Bowman[37] indicate that, for esters, the major radiolysis products are CO and CO_2. For aromatic esters the G ($CO+CO_2$) values reported by Kircher and Bowman range from 0.3 to 3.6 molecules per 100 eV. Based upon this cursory survey of the literature and the lack of any specific information on the organics actually contained in the Pathfinder waste packages, a G of 3.6 molecules per 100 eV will be assumed. This is the reported G-value for dibenzyl adipate. Thus, an aromatic compound with a particularly high G-value for carbon-containing gas production has been taken as the C-14-labeled substrate. (It should be noted that the actual effective G-value for the package could be lower than this value.) It is further assumed that C-14 is released along with C-12 in proportion to the atom ratio of these two isotopes in the aromatic compound; the aromatics have been assumed to be singly-labeled.

In one year, the total amount of energy given off through β^- decay in one millimole of the waste is 7.03×10^{20}eV (average β^- energy for C-14 taken as 49 keV). The G-value for carbon-containing gas production is 3.6 molecules per 100 eV absorbed and the atom fraction of C-14 to C-12 in singly-labeled dibenzyl adipate of specific activity 12.5 Ci/mole is 0.011. These values lead to an initial C-14 release of 2.8×10^{19} molecules CO_2+CO or 29 mCi per year per mole of substrate. From the specific activity given, 12.5 mCi/mmole, this means that the first year gas release from the Pathfinder Laboratory wastes is 56 mCi to Trench 14A, 13 mCi to Trench 23, and 38 mCi to Trench 24.

4.5.2.2 Combined Radiolysis and Simple Contact-Loss Leaching

The treatment of leaching due to simple loss on contact, which was applied to the generalized waste in Section 4.5.1.3, is also applicable to the waste in shipments >1 Ci.

A combined model for radiolysis and simple contact-with-water leaching has been developed in which the source term available for radiolysis and leaching is diminished simultaneously by these two mechanisms (in addition to radioactive decay). The 15% 55-gallon drum (breached on emplacement) factor has not been used because of the small number of drums involved. It must be remembered that part of the leaching model involves a five-year induction period to first pitting of the 55-gallon drums. There is therefore no leaching loss in the first five years, during which time only radiolysis losses occur. Radiolysis losses continuously decrease as the source term is depleted by the leaching mechanism. Leaching losses are seen to continuously increase as the time increases; this is a consequence of the steadily growing pit size in the drum through which leaching releases occur. Thus in the early years, radiolysis losses dominate and after a certain period, leaching losses dominate.

Table 4.13 gives the annual incremental releases of C-14 to Trenches 2, 14A, 23, and 24 for this model. As can be seen in the first three columns, the releases prior to, and including, the fifth year correspond only to gaseous losses of radiolysis products. The 55-gal drums have been assumed to present no barrier to gas release. By the tenth year, the radiolysis and simple leaching losses are of similar magnitude but by year 30, the leaching losses have become dominant. This behavior would be expected since both the radiolysis and leaching losses are dependent on the activity source and this is constantly being depleted by releases from both types of mechanism. However, the leaching losses, which are intimately connected to the amount of corrosion which has occurred (since this has been taken to limit the amount of water which is allowed contact with waste), steadily increase as the drums corrode away. Radiolysis losses, on the other hand, are dependent on the corrosion of the drum only in that it affects (through leaching) the depletion of the activity source. These radiolysis releases must, then, decrease with time.

Table 4.13

IFRs and Annual Incremental Releases in mCi of C-14 to Trenches 2, 14A, and 24 From >1 Ci Shipments - Corrected for Decay - Radiolysis and Simple Contact Leaching

Incremental Fraction Release: Year:	1	3	5	10	20	30	40	50	60	70	80	90
Radiolysis IFR	2.1 E-3	2.1 E-3	2.1 E-3	2.1 E-3	2.0 E-3	1.8 E-3	1.6 E-3	1.4 E-3	1.1 E-3	8.0 E-4	4. -4	6.0 E-5
2 (23000)[a]	48	48	48	48	46	41	37	32	25	18	10	1
14A (27000)	57	57	57	57	54	49	43	38	30	22	12	2
23 (12000)	25	25	25	25	24	22	19	17	13	10	5	1
24 (18000)	38	38	38	38	36	32	29	25	20	14	8	1
Simple Leaching IFR	0[b]	0[b]	0[b]	1.2 E-3	3.8 E-3	6.2 E-3	8.5 E-3	1.1 E-2	1.3 E-2	1.5 E-2	1.7 E-2	1.9 E-2
2 (23000)	0	0	0	28	87	143	196	253	299	345	391	437
14A (27000)	0	0	0	32	103	167	230	297	351	405	459	513
23 (12000)	0	0	0	14	46	74	102	132	156	180	204	228
24 (18000)	0	0	0	22	68	112	153	198	234	270	306	342
Total Radiolysis and Leaching IFR	2.1 E-3	2.1 E-3	2.1 E-3	3.3 E-3	5.7 E-3	8.0 E-3	1.0 E-2	1.2 E-2	1.4 E-2	1.6 E-2	1.7 E-2	1.9 E-2
2 (23000)	48	48	48	76	131	184	230	276	322	368	391	438
14A (27000)	57	57	57	89	154	216	270	324	378	432	459	515
23 (12000)	25	25	25	40	68	96	120	144	168	192	204	229
24 (18000)	38	38	38	59	103	144	180	216	252	288	306	343

[a]Initial trench activity in >1 Ci shipments in (mCi).
[b]55-gal drums (assumed all intact) given a 5-yr induction period to first pitting.

4.5.2.3 "Volume Removal" Model

In this model, it is assumed that the 55-gal drum corrosion limits the access of water to the waste materials and also that the activity releases from the drums are equivalent in volume to the water volume allowed to reach the wastes. The C-14 activity is assumed to be homogeneously distributed throughout the container and to have a uniform "activity density." Also, there is assumed to be no inhibition of removal of material, i.e., once the water has been allowed in the drum, it is taken to be able to exit freely, carrying with it, or rather displacing, an equivalent volume of waste material.

The amount of radioactive material to be released in a given time period according to this model is given by

$$dA = a(t) \cdot Q \cdot \rho \cdot dt \tag{4.29}$$

where

dA is the incremental amount of material released at time t (in mCi),

a(t) is the area of corrosion (cm^2) in the 55-gal drum at time t,

Q is the rainfall per year (89 cm/yr),

ρ is the activity density of the wastes (mCi/cm^3),

dt is the time interval of concern (yr).

The corrosion derivation is performed in Appendix C; the area of corrosion has been taken as:

$$a(t) = \pi k^2 t^2 \tag{4.30}$$

where

k is a corrosion constant relating to the burial site conditions

The assumptions involved in the substitution are discussed in Appendix C. Substitution in the formula for dA yields, on integration,

$$A = \frac{Q \cdot \rho \cdot \pi k^2 t^3}{3} \Bigg]_{t_1}^{t_2} \tag{4.31}$$

which has the form

$$A = Bt^3 \Big]_{t_1}^{t_2}$$

where B is 0.48 for Trench 2 and 1.25 for Trenches 14A, 23, and 24.

For example, for Trench 2:

$$\frac{Q}{3} \cdot \rho \cdot \pi \cdot k^2 = \frac{(89 \text{ cm/yr})\ (5.47 \text{ E-3 mCi/cm}^3)}{3}\ \pi(0.96)^2 = 0.48$$

Here, ρ, the activity density, has been calculated for a total of 23,000 mCi of C-14 waste being distributed over 20 total packages (each assumed to have a 55-gal, or 2.10×10^5 cm^3 capacity).

Results of the analysis are given in Table 4.14. It is seen that the only release for the first five years is the first year total loss from the 12 non-drum packages in Trench 2. The C-14 releases for the tenth year are identical for Trenches 14, 23, and 24 because the shipments in these trenches were such that the activity density had the same value for all three. Inherent in this model is the fact that the releases depend on the activity density, and not on the absolute total activities present. It is to be expected, whether the "simple leaching" model or the "volume release" model is used for the C-14 >1 Ci shipments, that radiolysis losses will dominate the releases for the first five years due to the assumption of a 5-year induction period to first pitting for the 55-gal drums. By year ten, as can be seen from comparison of Tables 4.13 and 4.14, the volume release model releases are the largest and their magnitudes remain higher throughout the time of interest. At some time in the years indicated in Table 4.14 (41 years for Trench 2, 33 years for Trench 14A, 26 years for Trench 23, and 29 years for Trench 24) the entire waste activity will have been depleted for these wastes, given the "volume release" model treatment.

Table 4.14

Annual Release (in mCi) of C-14 to
Trenches 2, 14A, 23, and 24 From >1 Ci Shipments
Volume Removal Model

	1	3	5	10	20	30	40
Trench							
2 (23000)[c]	0[a] 13,600[b]	0[a]	0[a]	29	303	865	1714 (41 yr all released)
14A (27000)[c]	0	0	0	76	789	3264 (33 yr all released)	
23 (12000)[c]	0	0	0	76	789 (26 yr all released)		
24 (18000)[c]	0	0	0	76	789 (29 yr all released)		

[a]55-gal drums were given a 5-yr induction period to first pitting.
[b]Release from 12 packages known not to be 55-gal drums.
[c]Initial total C-14 activity (mCi) from >1 Ci shipments in trench.

4.6 Release of I-129

The six shipments of I-129 to the trenches covered by this study have been considered in Section 2.3.2.3 and 2.4.2.3, and the information is summarized in Table 2.16. As discussed in Section 2.3.2.3, the shipments from all generators except the University of Illinois almost certainly contained only μCi amounts of I-129. The 1 mCi shipment from University of Illinois thus completely dominated the known site inventory.

Before the RSR of the 1 mCi shipment was found, the three other generators (the fourth, Atomic Disposal Co. was no longer in business) had been contacted regarding their waste form and packaging. Personnel from these companies stated that their waste was all in the form of calibration sources. One type of source described was made by applying a relatively thin layer of epoxy resin containing the I-129 as NaI, on a plastic backing. The other sources are assumed to have been similar. The University of Illinois material, on the other hand, was not in the form of sources. Some of it had been used as targets in nuclear physics experiments, and the waste was said to be in the form of used targets and unused material in the form purchased from the supplier. Thus, both used targets and unused material presumably had the chemical form of inorganic iodide, most likely NaI.

All shipments were made in standard 55-gal steel drums. Primary containment is not known for most of the shipments, including that from University of Illinois. Glass bottles and plastic (e.g., polyethylene) bags are the most likely containers, and neither can be considered to provide a significant barrier to water contact. Once water contacted the waste, release of I-129 would be rapid. Leaching of water-soluble salts (e..g, NaI) out of the targets and epoxy sources would be rapid, and the unused material in the 1 mCi shipment would dissolve at first contact with water. Two polar cases may therefore be considered for the release of I-129 - either all seven drums containing I-129 were among the 15% breached on emplacement, or none of them were. In the first case, all the I-129 would be released during the wetting fronts occurring in the first year after burial. In the second case, no release would occur until breaching of the drums, assumed to be in the fifth year after burial and all the I-129 would be released in a relatively short period after breaching. Release from the actual drums would presumably be somewhere between these two extreme cases.

4.7 Non-Fuel Cycle Sr-90, Cs-137, and Co-60

4.7.1 3M Co. Sr-90 and Cs-137

The inventories of Sr-90 and Cs-137 in Trenches 1 and 2 from 3M Co. shipments have been given in Table 4.2. Through contact with 3M staff, the composition of the wastes has been learned (see Memo to File From D. R. MacKenzie, June 16, 1983, reproduced in Appendix B). In general, the wastes were all from 3M hot cell operations and consisted of glass, paper, plastic, and some metal on which contamination was in the form of cesium and strontium as chloride, nitrate, and labelled microspheres in an activity ratio of

1/3:1/3:1/3. For the purposes of the release model, the cesium and strontium chlorides and nitrates have been assumed to be readily soluble and, thus, to be released on contact with water.

In a discussion with 3M personnel about leach rates from 3M-labelled microspheres, it was learned that leach test failure of the microspheres is often due to removable surface contamination. Additionally, there is, apparently, no certain way to estimate the amounts of "leach-test-passing" and "leach-test-failing" microspheres that might be in 3M hot cell wastes. Thus, it is quite likely that much of the microsphere waste could release activity on contact with water. The conservative assumption has been made that the microsphere activity is released at the same rate as that from the cesium and strontium salts.

4.7.1.1 Considerations for Model Development

The first aspect of a leach model addressed is the integrity of the lead-lined drum container (see Figure 4.1 for structural details). Corrosion of the 3-in. thick lead lining has been discussed in Section 4.2.2, and it was concluded that this corrosion did not negatively influence the ability of the lead lining to hold the waste material. Corrosion of the steel drum, however, must be considered in some detail. It has been noted[28] that, for carbon steel in the presence of lead, a galvanic couple is set up between the carbon steel and the lead which leads to an acceleration of the corrosion of the carbon steel. In other words, the lead is essentially given an extension of "protection" at the expense of the carbon steel.

Given that the most likely means by which the cesium and strontium activity could be released from the 3M packages is through leaching, the main determining factor in the rate of release for this material would be the rate and amount of contact with water. From Figure 4.1, it can be seen that water contact with the wastes is limited by the presence of the outer carbon steel drum. In this radionuclide release model, the corrosion of the carbon steel outer drum has been given a five-year "induction" period and then an additional interval during which corrosion will have proceeded to the extent that an area equivalent to that of a drum top will have corroded away. It is being assumed that, as a worst case, corrosion could occur in one concentrated area of significance with respect to the influx of water. The most obvious such "significant" area is the drum top area, since it is the drum top which has the most influence in inhibiting the flow of rainwater into the circular "crack" around the lid of the lead lining. In this model, it has thus been assumed that, by the time enough corrosion of the outer carbon steel has occurred to be equivalent to the drum top area, there is no longer any inhibition of access to the inside of the lead lining for the rainwater. The time necessary to corrode an area of carbon steel equivalent to the drum top area can be calculated from the corrosion equation:

$$S = ct^{1/2} \qquad (4.32)$$

where

S = corrosion pit area (cm^2)

c = corrosion rate constant for Sheffield site conditions and carbon steel drum ($cm^2 \cdot yr^{-1/2}$)

and,

t = time (yr).

Given that, for carbon steel drums in soil like that at the Sheffield site, an average drum has an 86-yr "lifetime" (to total corrosion), a ratio can be set up as follows:

$$\frac{S}{S_o} = \frac{t^{1/2}}{(86\ y)^{1/2}} \qquad (4.33)$$

where S_o is the total drum area and all the other variables have the same meaning. For a 30-gal carbon steel drum, the value for the ratio of drum top area to total area is 0.16 and substitution of this for S/S_o leads to a period of 2.3 yrs for corrosion of an area of carbon steel drum equivalent to the drum top area. Hence, there is a period of 7 yrs (5-yr "induction" plus 2 yr corrosion) during which it is assumed no leaching releases occur.

The modeled radionuclide releases from the 3M lead-lined drums will depend on the amount of rainfall assumed to contact the drums. Three separate cases of yearly rainfall have been considered: (1) rainfall I corresponds to that at the Sheffield ground surface [89 cm/yr[3]], (2) rainfall II (48.3 cm/yr) corresponds to the average of the rainfall at the ground surface and that which reaches the water table (6.35 cm/yr[3]), and (3) rainfall III corresponds to the latter, i.e., that which reaches the Sheffield water table (6.35 cm/yr[3]). Annual incremental radionuclide releases have been calculated for each of the three rainfall cases.

Another factor to be considered in model development is whether a diffusion-limited or a non-diffusion-limited approach is more appropriate. Two separate cases of loss or release by diffusion from a stirred solution of limited volume[38] were considered as potential models of the type of release to be expected in the trench environment for the lead-lined waste package.

Both of the diffusion-limited models yield releases that are smaller than those obtained with a non-diffision-limited total mixing "rinse" model.* It was decided that, since the former models could not effectively represent any drum positional orientation other than "upright", and, since it is known

*Details of the calculation are given in a memo to file, "Model for Releases of Cs-137 and Sr-90 from Wastes Shipped by 3M Co. Buried at Sheffield Site, 1967-1971, Trenches 1 and 2," by C. R. Kempf, December 29, 1983.

that there is no guarantee the drums have any one positional orientation in the trench, the "rinsing" model would be used. It can be relied upon to yield radionuclide release values more conservative (i.e., higher) than the diffusion-limited cases, and it can be modified to accommodate essentially any drum orientation. It is described in the following section.

4.7.1.2 "Rinse" Model

In this model, it was assumed that total mixing of the waste isotopes occurred subsequent to an "induction" period during which the rainwater which manages to enter the interior of the lead lining accumulates until it has reached the level at which it can exit the container. In other words, the container contents become a solution of soluble waste material in which there also are solid, insoluble waste components such as glass, plastic, and metal. The length of the "induction" period will necessarily depend on: (1) the rainfall case (I, II, or III), and (2) the positional orientation (angle) of the drum. The waste itself was considered similar to a known volume of a homogeneous mixture of radioisotopes "in solution." Instead of the radionuclide release occurring by diffusion out through the opening around the lining lid and then into the rainwater, the rainwater has been assumed to enter the drum, become homogeneous with the waste solution, and then exit the drum, carrying with it an amount of waste radionuclide equivalent to the simple product of the waste "solution concentration" and the rainwater volume. With respect to volumes, length of rainfall accumulation, etc., the scheme has been based on yearly totals. The model could be characterized as a type of rinse scheme in which the wastes are annually rinsed by a volume dependent on the rainfall, and in this rinse are acquired radionuclides which are then released to the trench environment.

In this model, the amount of radionuclide release is dependent on the relationship between two main factors, given any particular waste radionuclide "concentration". The factors are: (a) the volume of the rinse and, (b) the volume of the standing solution of waste radionuclides contained in the lead lining. The ratio of these two volumes, (a)/(b), has been called "z" in the derivation of the expression for the incremental package radionuclide releases. The steps of this derivation are outlined in Appendix E. Appendix E also contains calculations of various parameters specific to the model. The final expression for the activity incremental fraction release is:

$$IFR = z(1-z)^{n-1} \qquad (4.34)$$

where

IFR is the incremental fraction release,

z is the ratio of rinse volume to standing bath volume ($\ell/0$ in the derivation in Appendix E), and

n is the number of the rinse

Since the model assumes one rinse per year, successive values of n correspond to successive years, from the time rinsing begins.

Calculations of annual incremental and cumulative releases of Sr-90 and Cs-137 to Trenches 1 and 2 have been carried out for a period of 300 yrs. Fractional releases were first calculated, using the rainfalls, distribution of drum positions, and induction periods discussed in the text and in Appendix E. These values were then multiplied by the trench inventories to obtain the annual releases. These annual releases are given, on fiche, in Appendix D as Table D.5. An abbreviated table is also presented in Section 4.10.

4.7.1.3 Effects of Drum Orientation

A. "Rinse" Model Drum Orientations - 0° to 90°, Inclusive

As would be expected, both the volume of the rinse, (a), and the volume of the standing bath, (b), depend on the positional (angular) orientation of the waste drum in the trench. For example, the volume of the rinse to which an upright waste drum is subjected is simply the product of the height of a column of rainfall (rainfall I, II, or III) times the area available for water influx (the area of the opening around the lead lining lid). A drum at a 45° angle to upright presents a smaller area to (assumed) downward rainfall than an upright drum, i.e., for the drum at 45°, the rainwater influx area has become the 45° projection of the upright drum opening area and its value is cos 45° times the upright drum opening area.

Similarly, an upright drum will be expected to have the largest standing bath volume (equivalent to the free space of the drum interior) while a drum at a 45° angle to upright will have a standing bath volume that has been calculated to be 75% of that for the upright drum. This is illustrated in the diagrams of Figure 4.2.

The radionuclide releases have been calculated for several drum positional orientations: upright, 30°, 45°, 60°, and 90° to upright. In all but the last case, the rainfall influx area is calculated as (lid opening area) times cos(angle). The 90° to upright case was treated such that the rinse volume was fixed by rainfall through 50% of the upright lid opening area, and the standing bath volume was taken as 50% of the available internal drum free space (both the rinse volume and the standing bath volume were based on the assumption that rainfall entrance and rinse exit areas were equivalent in size). The "30° to upright" case yielded the highest radionuclide incremental fraction releases, and it was taken as the most conservative waste package representation. Thus, any drum orientation ranging from the "upright" position to the "90° to upright" case has been taken as having the same incremental fraction releases as the "30° to upright" orientation.

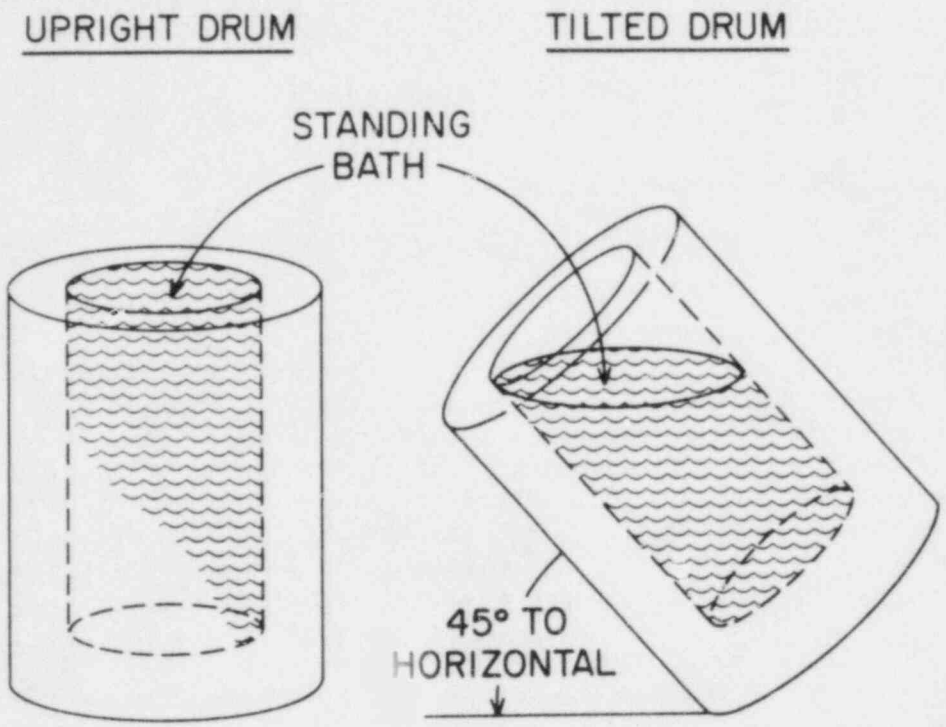

Figure 4.2 Water standing in upright and tilted lead-lined drums. For simplicity the lids are not shown.

B. "Wash" Model Drum Orientations -- >90° to 180°

It is equally likely that the 3M lead-lined drums could have positional orientations at angles greater than 90° up to the totally upside down position, i.e., 180° to upright. By symmetry, the angles from 180° to the full 360° would already be covered by consideration of the 0° to 180° range. Once the lead-lined drum angle exceeds the horizontal (90°) position, there is a finite probability that the lead lid may dislodge from the top of the lining cylinder and essentially cause the lead lining portion of the container to open. For the purposes of modeling the radionuclide releases from these packages, the worst case of a dislodged lid, physically displaced from the lining cylinder by 1.75 in. (the clearance between the inner lead lining lid and outer carbon steel lid), has been used as the physical

representation of the waste package. Implicit in this worst case picture are the assumptions that (1) the carbon steel outer drum corrodes such that the area of the lid is affected first and most seriously, and thus allows nearly uninhibited water influx and, (2) the inner lead lining is adherent to the outer carbon steel shell and the latter is sufficiently strong that the lead lining cylinder is physically held in place above the lining lid, i.e., prevented from falling on top of the dislodged lid. The outer carbon steel drum has thus been idealized as totally porous (water influx allowed) and yet strong (supporting the lower part of the lead lining).

The radionuclide release from drums in the dislodged lid condition has been modeled such that the amount of released material is taken as equivalent in volume to the amount of water that contacts the wastes, i.e., for a rainwater influx of 500 cm^3, 500 cm^3 of waste material (and thus, the assumed homogeneously distributed radionuclides) would be released. This is essentially a type of "wash" mechanism, and it is assumed that the waste materials are compacted in the lead-lined drums in such a manner that there is annual replenishment of waste to the region of water influx to the drum (i.e., to that area around the lid). This type of release mechanism model is, like the 0^o to 90^o drum waste release mechanism model, dependent on the amount of rainfall, and the same three rainfall cases (I, II, and III) have been considered.

C. Distribution of Drum Positions

It has been assumed that the drums are distributed in totally random physical orientations in the trenches. The angular drum position possibilities can be described, to be consistent with the sequence of consideration involved in the release model development, as spread over five orientations:

(1) upright,

(2) tilted at some angle between upright (0^o) and horizontal (90^o),

(3) horizontal (90^o),

(4) tilted at some angle between horizontal (90^o) and upside down (180^o), and

(5) upside down.

As has been discussed, there are two main mechanisms that have been used for modeling the radionuclide release from these lead-lined packages: (a) the "rinse" mechanism model which has been applied equally for positional orientations (1), (2), and (3); and (b) the "wash" mechanism model which has been

applied to positions (4) and (5). Given five possibilities for the drum position in the trench and given that three of the five orientations are modeled by the "rinse" model, the source term for the total available radionuclide concentration (Cs-137 and Sr-90) in these 3M wastes has been distributed as 60% requiring the "rinse" release mechanism (3 out of 5 = 60%) and 40% requiring the "wash" or upside-down-dislodged-lid release mechanism (2 out of 5 = 40%).

D. Vertical Distribution of Drums

The three separate rainfall cases that have been considered in the modeling of release from the 3M lead-lined packages could be considered to correspond to different positions in the trench, i.e., the highest rainfall, (I), might correspond to that experienced by a package in a position near the top of the trench while rainfall II might represent that experienced by a package near the middle, and rainfall III that experienced by a package near the lowest part of a trench. Since it is not known where, with respect to elevation, these 3M packages were emplaced in the trenches, it has been assumed that they occupy random positions and are thus equally distributed over the trench elevation. The model incremental fraction releases from these packages are therefore distributed equally over the three rainfall cases (i.e., 1/3 subjected to rainfall I, 1/3 subjected to rainfall II, and 1/3 subjected to rainfall III).

4.7.1.4 Overall Profile of Releases

For the 60% of activity that has been given the "upright" drum, rinse model treatment, it is expected that there will be no release until after the 5-yr outer drum corrosion induction period plus the 2-yr drum top disintegration period plus the respective induction periods during which the rainwater accumulates in the standing bath. For the highest rainfall (rainfall I), the standing bath accumulation takes 8 years and thus the first release should not occur until after the fourteenth year, or beginning in the fifthteenth year. The outline of release for these drums should build up after the fifthteenth year, reach a maximum and then gradually decrease as both the rinsing loss and decay loss contribute to the diminution of the waste activity source.

The 40% of the activity that is subjected to the "wash" release model, can be expected to start releases in the seventh year and to reach the maximum release quickly since the wash mechanism results in quite rapid total loss (following "induction": rainfall I, first year total loss; rainfall II, total loss by the second year; and rainfall III, total loss by the fourth year).

4.7.2 ANL Sr-90 and Cs-137

On the basis of the factors discussed in Section 4.2.3, it is not expected that the aluminum MAP tubes used to contain the relatively high-specific activity waste in ANL's special series of shipments in 1970 would be readily penetrated by corrosion. However, they were closed by a press-fit

lid, and water could enter through the space between the lid and the tube. Depending on the tightness of fit of the lid, the rate at which water would enter might be quite low, particularly given the intermittent nature of wetting fronts in the trench. The angle at which the tube rested is also an important factor. Those at an angle near the horizontal would likely be more apt to have water enter during a wetting front than those more nearly vertical. In the case of upright tubes, water flowing past the lid would not develop much of a head to force its way through the narrow channel between lid and tube. In the case of upside down tubes, when water collected around the lids (now resting on what was the top of a drum), it would be forced up into the tubes only a relatively short distance because of the pressure of the trapped air. In tubes nearer the horizontal, some of this air could escape and water could fill most of the tube volume if sufficient had collected in the outer (drum) container.

The drums, of course, might never fill, especially after the 5-year-induction period for first penetration of initially unbreached drums. As more holes appear in a drum, there is more chance that water will merely move through it without collecting in it to an appreciable extent. Hence, a MAP tube would not necessarily ever have had its lid portion standing in water. Once the trench soil settled around the tubes as a drum became progressively more corroded, wet soil would be in contact with the lids during a wetting front, but here again, only a small amount of water might enter the tube, depending on its angle of rest. All things considered, there is much less access to water in the case of these tubes than in the case of the lead-lined drums discussed in Sections 4.2.2 and 4.7.1 However, it must be expected that some tubes would have water enter in a time of concern for activity release.

The most important factor in this release appears to be the leaching rates of the waste. Leach rates of fission products and TRU isotopes from irradiated UO_2 fuel in deionized water and river water have been determined by Vandergraaf.[39] His results are in line with those of earlier work[40] with UO_2 and are used in this assessment. They showed that, after 900 days, Cs was the only element which displayed significantly greater proportional release from the UO_2 than the matrix element itself, and this was about a factor of 10 greater. The matrix dissolution rate was approximately 1×10^{-6} g/cm^2-d, using the conservative assumption that the actual surface area of the samples was the geometrically measured area. The releases, as percentage of fission product inventory, were initially $>10^{-4}$% per day, but by 900 days had dropped by an order of magnitude.

In terms of the ANL waste, these results indicate that Cs-137 and Sr-90 release would not be rapid when the waste was subject to leaching. The rate would be somewhat greater than the fractional release rate indicated by Vandergraaf's study ($<10^{-6}$ per day for Sr-90 and $<10^{-5}$ per day for Cs-137) because of the small particle size of the waste. When it is borne in mind that the ANL waste would see water only intermittently, the annual release rates will still be quite low even for small particles. Using the conservative matrix dissolution rate of 10^{-6} g/cm^2-d given by Vandergraaf, the time required for complete solution of a spherical particle of 0.1-mm radius is approximately 3×10^4 days or over 600 years, assuming Sheffield trench

water behaved the same way as Vandergraaf's river water and intermittent wetting caused the same amount of leaching as that in the experiments (static leaching with frequent water changes). Thus, since the Sr release rate is essentially the same as that for uranium, the UO_2 particle of 0.1 mm radius would not release all its Sr for several hundred years, and Cs would be released in one tenth the time taken for the Sr.

The particle size distribution of ANL's UO_2 waste is, of course, not known. However, given the leach rates just discussed, it seems likely that releases of Cs-137 and Sr-90 would be minor compared to that from the 3M waste, and probably insignificant. In the case of the 1970 special series of drum shipments, where the waste was contained in the aluminum MAP tubes discussed in Section 4.2.3, this is particularly so, since it is uncertain whether much of the waste would be contacted with water in times short enough to be of concern. Even in the case of the 1968 series of special bin shipments, where the waste would be expected to be exposed within a relatively short time (<10 years), it is likely that absolute releases would be minor. Although there is no uncertainty about exposure of the latter waste to water, the low UO_2 leaching rates and the relatively small absolute amounts of the waste support this position. Thus, we have chosen not to include (probably minor) releases of the ANL waste in the overall release data for Trenches 1 and 2.

4.7.3 Co-60 From ANL and University of Wisconsin

The University of Wisconsin was the only non-fuel cycle generator to ship a reasonably large amount (84 Ci to Trench 1) of Co-60 by itself. The only other sizeable amounts, which are only rough estimates, were in waste associated with irradiated fuel, sent in ANL special series of shipments, and have been assigned values of 5 Ci for Trench 1 and 86 Ci for Trench 2.

The non-fuel cycle contributions to Trenches 1 and 2 are small compared with the estimated fuel cycle contributions of approximately 700 and 520 Ci, respectively. The University of Wisconsin shipments of 80 and 4 Ci were each Co-60 irradiators; the smaller one, at least, was of a kind that was commercially available. Both almost certainly consisted of irradiated cobalt metal encapsulated in another metal, most likely stainless steel. The ANL Co-60 would have been formed by irradiation of the stainless steel cladding and other stainless steel components of fuel rod assemblies, and as such would have been essentially uniformly distributed throughout the steel. Most of the steel was presumably in massive form, but some would be in the form of small particles from cutting fuel rods, etc. The amount in particle form would be expected to be only a small fraction of the total.

Release of Co-60 from the irradiators could not begin until the stainless steel encapsulating the cobalt had been penetrated by corrosion, and then would be slow due to the low dissolution rate of cobalt metal (via corrosion). In the case of the ANL waste, release of Co-60 would begin as soon as water contacted the stainless steel and started corroding it, but would be very slow because of the low corrosion rate of stainless steel. As a result of these considerations and the relatively small amount of non-fuel cycle Co-60, its

contributions to the total release of Co-60 from Trenches 1 and 2 will be small, and almost certainly insignificant.

4.8 Release of Co-60, Cs-137, and Sr-90 From Fuel Cycle Waste

4.8.1 Isotope Inventory in Different Package-Waste Form Combinations

To model the releases of Co-60, Cs-137, and Sr-90 from the fuel-cycle waste to the Sheffield trenches of concern, required identification of major release modes and partitioning the total inventory of these isotopes among the release modes. The approach taken was to assemble a detailed inventory of the content and characteristics of the fuel-cycle wastes emplaced in Trench 24. This trench was chosen for detailed examination because it received the largest amount of fuel-cycle waste containing relevant isotopes of any trench examined in this study (see Table 2.33). It was also filled near the midpoint of Sheffield's operating life. Therefore, it is assumed to be the most representative single trench in regard to fuel cycle wastes. The detailed examination amounted to characterizing the waste shipped by the major waste generators. This was accomplished by either obtaining the information directly from the RSRs or, more frequently, from direct contact with the waste shipper.

The major fuel-cycle shippers in decreasing order of the total curie contribution were: Commonwealth Edison, Monticello, Nebraska Public Power District (NPPD), Anefco and ATCOR (both brokers), Northern States, and Consolidated Edison. Based on the descriptions that could be obtained, the waste was partitioned into six generic categories. These are tabulated in Table 4.15 along with their contributions to the volume and to the activity of fuel cycle waste in Trench 24.

Table 4.15

Partitioning of Fuel-Cycle Waste in Trench 24

Type/Packaging	% Total Volume[a]	% Total Activity[b]	Relative Partitioning[c]		
			Mont.	Comm. Ed.	Remainder
cement solidified/drum	56	55		82	33
cement solidified/liners	12	11		18	0.2
UF/drum	3	1	3		
UF/liners	7	23	97		
misc unsolidified/drum	6	6		0.2	37
misc unsolidified/liners	17	5			30

[a]Based upon total fuel-cycle volume in Trench 24 contributed by the major generators.
[b]Based upon total fuel-cycle activity in Trench 24 contributed by the major generators.
[c]The relative activity distributions (in percent) contributed to Trench 24 by these fuel-cycle generators. Remainder denotes the contribution of all other major generators except Monticello (Mont.) and Commonwealth Edison (Comm. Ed.).

The majority of waste disposed of in Trench 24 was cement solidified in 55-gal drums. This was primarily shipped by Commonwealth Edison with much smaller contributions from NPPD and Consumer Power. The second largest contribution on the basis of activity was the liners of urea-formaldehyde (UF) solidified waste shipped exclusively by Monticello. Smaller contributions resulted from the other categories listed in the table. Since all of the UF solidified waste was shipped by Monticello, the modeling inventory figures for UF were taken from Tables 2.31 and 2.32.

The balance of activity of Co-60 and Cs-137 (all shipments minus Monticello, see Table 2.31) were partitioned among the remaining four categories. This task was far more difficult than the partitioning of activities of the U-F solidified waste. This was due to the number of remaining major shippers (seven), the variety of containers (boxes, drums with volumes from 30 to 55-gal, and liners with volumes from 50 to 400 ft^3), and the character of the waste itself. The total inventory of Sr-90 (see Table 2.32) was partitioned among all waste types on the basis of Cs activity (see Section 2.3.3.5). In general, cement solidified waste includes wet wastes, such as boric acid and sodium sulfate concentrates, and demineralizer resins. The overwhelming majority of drums were 55-gal in volume. Several shipments of 30-gal drums (111 drums containing 2.2 Ci, Consumer Power) and small 50-ft^3 liners (1.1 Ci in 33 liners, Northern States) were also included in this category. Liners of cement solidified waste were shipped solely by Commonwealth Edison (CE). Of the total 172 CE liners shipped to Trench 24, 163 were 112 ft^3.

Miscellaneous waste included all other unsolidified wastes. These wastes would range from dry-active waste to dewatered demineralizer resins. The decision to include dewatered resins with miscellaneous waste was based on the leach data of Colombo,[41] which suggested rapid release of the radioactivity by leachants with high conductivity. This situation should occur based on the data presented in Reference 41. The liners of miscellaneous waste primarily shipped by Anefco and Actor, had an average volume of ~122 ft^3, but ranged from 50 to 400 ft^3. Drums were predominately 55 gal in volume.

Commonwealth Edison and Monticello wastes were treated separately because of their large contribution to Trench 24. All other generators were treated together. The relative partitioning of waste activity (see Table 4.15) obtained from the detailed examination of Trench 24 was subsequently applied to the inventories (see Tables 2.31 and 2.32) of Cs-137, Co-60, and Sr-90 of all trenches examined in this study. The results are shown in Table 4.16. The entries for UF waste result solely from Monticello. The entries for other categories are the sum of the contributions from Commonwealth Edison and all other generators. These values provide the input to the models described in the following section (4.8.2).

Table 4.16

Fuel Cycle Inventories of Co-60 and Cs-137[a] in the Selected Sheffield Trenches

	Trench	Activity (Ci)					
		In Drums				In Liners	
		Co-60		Cs-137		Co-60	Cs-137
cement solidified (wet wastes and resins)	1	262	(222/40)[b]	109	(93/16)	14[c]	15
	2	173	(147/26)	40	(34/6)	3	1.5
	7	35	(30/5)	28	(24/4)	5	6
	11	148	(126/22)	143	(121/22)	26	30
	14A	917	(779/138)	962	(818/144)	182	208
	23	823	(699/124)	829	(704/125)	156	177
	24	681	(579/102)	723	(615/108)	136	157
	25C	127	(108/19)	124	(105/19)	23	26
U-F solidified (wet wastes) and resins)	1	0		0		0	0
	2	0		0		0	0
	7	0.2	(0.17/0.03)	0.2	(0.17/0.03)	7.8	7.8
	11	4	(3.4/0.6)	4	(3.4/0.6)	154	154
	14A	3	(2.6/0.4)	3	(2.6/0.4)	104	104
	23	4	(3.4/0.6)	4	(3.4/0.6)	146	146
	24	9	(7.7/1.3)	9	(7.7/1.3)	328	328
	25C	0.01	(0.009/0.001)	0.01	(0.009/0.001)	0.49	0.49
misc. unsolidified (DAW, resin, etc.)	1	231	(197/35)	46	(39/7)	185	37
	2	192	(163/29)	39	(33/6)	153	31
	7	13	(11/2)	2.7	(2.3/0.4)	11	2
	11	32	(27/5)	6.6	(5.7/0.9)	25	5
	14A	104	(89/15)	22	(19/3)	82	16
	23	138	(117/21)	29	(25/4)	108	21
	24	64	(54/10)	13	(10/3)	49	9
	25C	25	(21/4)	4.8	(4.1/0.7)	20	4

[a]For the purpose of modeling Sr-90 releases, 1% of the Cs-137 values were used as the initial Sr-90 content (see Section 2.3.3.5).

[b]The values in parentheses are the amounts assigned to intact and failed drums at the time of emplacement. Based on data from the Hanford site, 15% of the drums are breached or dented at the time of burial. Dented drums are assumed to corrode very rapidly at stress points, and therefore, in effect, are breached at emplacement.

[c]Liners were assumed to be intact on emplacement.

4.8.2 Modeling of Isotopic Releases From the Fuel Cycle Waste

Given the lack of any experimental data pertinent to the release of radioactivity from a partially-breached container of solidified waste, any model attempting to describe such releases is at best speculative. Despite this, there are several features that such a model should possess. First, the presence of the outer container should retard the release of material from the solidified form. Initially, the container provides a complete barrier to water contacting the waste. Hence, at this time, non-gaseous release of radionuclides should be zero. Once breach of containment occurs, release via leaching can occur. Since the amount of material released to a leachate is dependent on the exposed surface area, the presence of a partially-corroded container ought to mitigate the releases. Secondly, releases from a partially-breached drum of solidified waste are most likely limited by the release from the waste solid. Thirdly, the effects of wet-dry cycling of the waste form would probably be to reduce the releases below that of continually-immersed waste. The latter point is supported by the work of Dayal et al.[42] Finally, leaching will occur over a time frame such that decay of the radionuclides is important.

To incorporate these features into a quantitative model for the prediction of the release of radionuclides from buried solidified fuel cycle waste at Sheffield, several assumptions had to be made. The first was that the leaching mechanism was diffusion controlled. This meant that release from the monolithic form could be described as a function of time and diffusivity. In particular, the cumulative fraction release, CFR, at any time, t, could be expressed by

$$CFR(t) = \sum_{n=0}^{i} C_n X(t)^n \qquad (4.35)$$

where

$$X(t) = \frac{S}{V}\sqrt{Dt}$$

where S is the surface area of the form, V its volume, and D the effective diffusivity of the radionuclide of interest. In the short term, Equation 4.31 is the familiar

$$CFR(t) = \frac{2S}{V}\sqrt{\frac{Dt}{\pi}} \qquad (4.36)$$

For the purposes of the model, however, the more realistic, infinite plane sheet solution given in ANS 16.1[34] was chosen. In this case, the data given in Figure 3 of ANS 16.1 was digitized and fit with a quadratic power series. Thus, Equation 4.1 becomes

$$CFR(t) = C_0 + C_1X + C_2X^2 \qquad (4.37)$$

where $C_0 = -0.0254$, $C_1 = 1.3441$, and $C_2 = -0.4416$. Since C_0 is negative, at short times, i.e. when $X < 0.019$, CFR would be <0. Thus, for the purposes of the model, C_0 was set equal to zero.

In order to describe the corrosion of the container, it was first necessary to obtain an estimate of the time to the formation of the first pit and the lifetime of the drum. In the case of cement solidified waste, these were estimated to be 4 yrs and 86 yrs, respectively, based upon the soil corrosion of mild steel data given by Romanoff.[13] For containers which contained waste solidified using urea formaldehyde, corrosion from within probably determines the container life. Using corrosion data from Colombo and Neilson[41] a time to pit initiation of 0.8 yrs and a drum life of 11 yrs were assumed. Once container lifetime estimates were obtained, it was necessary to describe the increase in exposed surface area as a function of time. That is, the fractional surface area exposed, A, had to be described. Several functions were considered: $A = kt^{1/2}$, $A = k't$, $A = k''t^2$. Because the first of these gives the largest increase in surface area in shorter times, it would lead to conservatively higher releases than the other two functions. This is because decay of the radionuclides of interest (Cs-137, Sr-90, Co-60) is appreciable over the container lifetime. Thus,

$$A(t) = S_o + (Kt)^{1/2} \qquad (4.38)$$

where S_o is the initial exposed surface area upon emplacement and K is a constant determined by the initial container area.

Once an effective surface area at any time, t, is obtained, then the cumulative fraction release at that time (not corrected for decay) may be expressed in terms of Equation 4.38 as

$$CFR(t) = \frac{A(t)}{S} [C_1X + C_2X^2] \qquad (4.39)$$

The incremental fraction release at t, IFR(t), is

$$IFR(t) = CFR(t) - CFR(t - 1) \qquad (4.40)$$

The affect of decay of the radionuclide of concern is accounted for by multiplying IFR by $e^{-\lambda t'}$ where t' is the time since emplacement. The decay corrected CFRs were obtained by summing the appropriate decay corrected IFRs.

In the discussion above, it is important to note that the leach time t is less than the decay time t'. This is for two reasons. First, there is the induction period to allow for breaching of the container. Second, once breaching occurs, the monolith is subject to only infrequent wetting. For the case of Sheffield, it was assumed, based on the number of rain days, that leaching only occurs for one-eighth of the time. Thus, for cement in a 55-gal drum, 300 yrs after emplacement, i.e., t' = 300, the total leach time is only 37 yrs.

The model was coded in FORTRAN and the resulting code listed in Table 4.17 was run for nine cases. These are:

1. Cement solidified waste in a 55-gal drum.
2. Cement solidified waste in a failed 55-gal drum -- 15% failure occurs during emplacement.
3. Cement solidified waste in a 100 ft^3 liner.
4. Urea-formaldehyde solidified waste in a 55-gal drum.
5. Urea-formaldehyde solidified waste in a failed 55-gal drum -- failure occurs during emplacement.
6. Urea-formaldehyde in a 100 ft^3 liner.
7. Unsolidified trash and resin waste in a 55-gal drum.
8. Unsolidified trash and resin waste in a failed 55-gal drum -- failure occurs during emplacement.
9. Unsolidified trash and resin waste in a 100 ft^3 liner.

Table 4.17

FORTRAN Code to Calculate the
Incremental and Cumulative Fractional Release From
Waste Solidified in a Carbon Steel Container

```
      PROGRAM SHEFLH(INPUT,OUTPUT,TAPE6,TAPE5)
      DIMENSION A(3,3000),CFR(3,3000),T(3,3000),DI(3),HL(3),
     1AR(3,300),EAD(10)
      REAL IFR(3,3000),LAMB
C       INPUT
    1 CONTINUE
      WRITE 40
      READ(5,45) RES
      IF(RES.EQ."QUIT ") GO TO 1000
      WRITE 5
    5 FORMAT(1X,"ENTER HEADING",/)
      READ(5,10) (EAD(NX),NX=1,8)
   10 FORMAT(8A10)
      WRITE 15
   15 FORMAT(1X,"ENTER  HALF LIFE IN YEARS:"
     1,/,1X,"[   CS   ][   SR   ][   CO   ]",/)
      READ(5,20) (HL(LH),LH=1,3)
   20 FORMAT(3G10.4)
      WRITE 22
   22 FORMAT(1X,"ENTER DIFFUSIVITY:",/,1X,
     1"[   CS   ][   SR   ][   CO   ]",/)
      READ(5,20) (DI(ID),ID=1,3)
      WRITE 25
   25 FORMAT(1X,"ENTER CONSTANTS ",
     1"FOR RELEASE EQUATION",/,1X,"[   CO   ]"
     2,"[   C1   ][   C2   ]",/)
      READ(5,30) C0,C1,C2
   30 FORMAT(4G10.4)
   40 FORMAT(1X,"ENTER ISOTOPE(A5):",/)
   45 FORMAT(A5)
C     CALCULATE FRACTIONAL SURFACE AREA AVAILABLE FOR LEACHING
      WRITE 100
  100 FORMAT(1X,"ENTER VOLUME,SURFACE AREA,INITIAL AVAILABLE",
     1"SURFACE AREA,",/,1X,"TIME TO FIRST PIT AND LIFE OF ",
     2"CONTAINER",/,1X,"[   V    ][   S    ][   SI   ][ TTP   ]",
     3"[   TL   ]",/)
      DO 110 I=1,3000
  110 A(I)=0.0
      READ(5,120) V,S,SI,TTP,TL
```

Table 4.17, Continued

FORTRAN Code to Calculate the
Incremental and Cumulative Fractional Release From
Waste Solidified in a Carbon Steel Container

```
  120 FORMAT(5G10.4)
      ITP=TTP*10
      ITL=TL*10
      CON=S/(TL**0.5)
      DO 130 I2=1,3000
      IF(I2.GE.ITP) GO TO 125
      A(I2)=SI
      GO TO 130
  125 A(I2)=SI+(CON*((0.1*I2)**0.5))
      IF(A(I2).GE.S) A(I2)=S
  130 A(I2)=A(I2)/S
C     CALCULATE CUMULATIVE AND INCREMENTAL RELEASES
      DO 400 IJ=1,3
      LAMB=ALOG(2.0)/HL(IJ)
      D=DI(IJ)
      DO 205 J=1,3000
      CFR(IJ,J)=0.0
  205 IFR(IJ,J)=0.0
      TC=0.0
      DO 250 J1=1,3000
      JM=J1-1
      T(J1)=0.1*J1
      IF(A(J1).EQ.0.0) GO TO 250
      TC=TC+(0.1/8.0)
      CFR(IJ,J1)=A(J1)*FR(TC,S,V,D,C0,C1,C2)
      IF(J1.EQ.1) IFR(IJ,J1)=CFR(IJ,J1)
      IF(J1.NE.1) IFR(IJ,J1)=CFR(IJ,J1)-CFR(IJ,JM)
      IF(IFR(IJ,J1).LE.0.0) IFR(IJ,J1)=0.0
  250 CONTINUE
      DO 260 K1=1,300
  260 AR(IJ,K1)=0.0
C     DECAY CORRECT CFR AND IFR
      DO 270 JD=1,3000
      DECAY=EXP(-0.1*JD*LAMB)
      IFR(IJ,JD)=IFR(IJ,JD)*DECAY
      JZ=JD-1
      IF(JD.EQ.1) CFR(IJ,JD)=IFR(IJ,JD)
      IF(JD.GT.1) CFR(IJ,JD)=CFR(IJ,JZ)+IFR(IJ,JD)
```

Table 4.17, Continued

FORTRAN Code to Calculate the
Incremental and Cumulative Fractional Release From
Waste Solidified in a Carbon Steel Container

```
  270 CONTINUE
C     CALCULATION OF ANNUAL RELEASES
      DO 300 K=10,3000,10
      KZ=K/10
      M=K-10
      IF(KZ.EQ.1) AR(IJ,KZ)=CFR(IJ,K)
      IF(KZ.GT.1) AR(IJ,KZ)=CFR(IJ,K)-CFR(IJ,M)
  300 CONTINUE
  400 CONTINUE
C     OUTPUT
      WRITE(6,500) (EAD(NZ),NZ=1,8)
  500 FORMAT(8A10)
      WRITE(6,510)
  510 FORMAT(20X,"CS",20X,"SR",20X,"CO",/,3X,
     1"TIME",3X,3(4X,"CFR",8X,"IFR",4X),/)
      DO 520 NO=10,3000,10
      NQ=NO/10
      WRITE(6,515) T(NO),(CFR(NU,NO),AR(NU,NQ),NU=1,3)
  515 FORMAT(1P,7G10.4)
  520 CONTINUE
      GO TO 1
 1000 CONTINUE
      STOP
      END
      FUNCTION FR(T,S,V,D,C0,C1,C2)
      X=((D*T)**0.5)*S/V
      FR=C0+(C1*X)+(C2*X*X)
      RETURN
      END
..
```

For the last three cases, the release function was modified to have the release at any time directly proportional to the surface area exposed at that time and to the fraction of the material remaining in the container at that time. This version of the code is listed in Table 4.18. The input parameter for each of these cases is given in Table 4.19. The diffusivities used for cases 1-6 were assumed to reflect the approximate order of magnitude values for the isotopes of concern in the two solidification agents. The values used are consistent with leaching data available in the literature.[41,43,44]

Once CFRs and IFRs were obtained for each of the nine cases, these values could be multiplied by the activity in each trench appropriate to a given case. These values were obtained by a review of the trench inventory data (see Table 4.16). The releases, by case, were then summed to give a total fuel cycle trench release. Both the fractional releases by case and the annual releases by trench are given on fiche in Appendix D, Tables D.3 and D.4, for Cs, Sr, and Co.

Table 4.18

FORTRAN Code to Calculate the Incremental and Cumulative Fractional Releases From Unsolidified Waste and Resin in a Carbon Steel Container

```
      PROGRAM SHEFLH(INPUT,OUTPUT,TAPE6,TAPE5)
      DIMENSION A(3,3000),CFR(3,3000),T(3,3000),DI(3),HL(3),
     1AR(3,300),EAD(10)
      REAL IFR(3,3000),LAMB
C       INPUT
    1 CONTINUE
      WRITE 40
      READ(5,45) RES
      IF(RES.EQ."QUIT ") GO TO 1000
      WRITE 5
    5 FORMAT(1X,"ENTER HEADING",/)
      READ(5,10) (EAD(NX),NX=1,8)
   10 FORMAT(8A10)
      WRITE 15
   15 FORMAT(1X,"ENTER  HALF LIFE IN YEARS:"
     1,/,1X,"[   CS   ][   SR   ][   CO   ]",/)
      READ(5,20) (HL(LH),LH=1,3)
   20 FORMAT(3G10.4)
      WRITE 22
   22 FORMAT(1X,"ENTER DIFFUSIVITY:",/,1X,
     1"[   CS   ][   SR   ][   CO   ]",/)
      READ(5,20) (DI(ID),ID=1,3)
      WRITE 25
   25 FORMAT(1X,"ENTER CONSTANTS ",
     1"FOR RELEASE EQUATION",/,1X,"[   CO   ]"
     2,"[   C1   ][   C2   ]",/)
      READ(5,30) C0,C1,C2
   30 FORMAT(4G10.4)
   40 FORMAT(1X,"ENTER ISOTOPE(A5):",/)
   45 FORMAT(A5)
C     CALCULATE FRACTIONAL SURFACE AREA AVAILABLE FOR LEACHING
      WRITE 100
  100 FORMAT(1X,"ENTER VOLUME,SURFACE AREA,INITIAL AVAILABLE",
     1"SURFACE AREA,",/,1X,"TIME TO FIRST PIT AND LIFE OF ",
     2"CONTAINER",/,1X,"[   V    ][   S    ][   SI   ][  TTP   ]",
     3"[   TL   ]",/)
      DO 110 I=1,3000
  110 A(I)=0.0
      READ(5,120) V,S,SI,TTP,TL
```

Table 4.18, Continued

FORTRAN Code to Calculate the Incremental and Cumulative Fractional Releases From Unsolidified Waste and Resin in a Carbon Steel Container

```
  120 FORMAT(5G10.4)
      ITP=TTP*10
      ITL=TL*10
      CON=S/(TL**0.5)
      DO 130 I2=1,3000
      IF(I2.GE.ITP) GO TO 125
      A(I2)=SI
      GO TO 130
  125 A(I2)=SI+(CON*((0.1*I2)**0.5))
      IF(A(I2).GE.S) A(I2)=S
  130 A(I2)=A(I2)/S
C     CALCULATE CUMULATIVE AND INCREMENTAL RELEASES
      DO 400 IJ=1,3
      LAMB=ALOG(2.0)/HL(IJ)
      D=DI(IJ)
      DO 205 J=1,3000
      CFR(IJ,J)=0.0
  205 IFR(IJ,J)=0.0
      TC=0.0
      DO 250 J1=10,3000,10
      JM=J1-10
      T(J1)=0.1*J1
      IF(A(J1).EQ.0.0) GO TO 250
      TC=TC+(0.1/8.0)
      IF(J1.EQ.10) IFR(IJ,J1)=A(J1)
      IF(J1.EQ.10) CFR(IJ,J1)=IFR(IJ,J1)
      JW=J1-10
      IF(J1.GT.10) IFR(IJ,J1)=A(J1)*(1.0-CFR(IJ,JW))
      IF(J1.GT.10) CFR(IJ,J1)=CFR(IJ,JW)+IFR(IJ,J1)
      IF(CFR(IJ,J1).GT.1.0) CFR(IJ,J1)=1.0
  250 CONTINUE
      DO 260 K1=1,300
  260 AR(IJ,K1)=0.0
C     DECAY CORRECT CFR AND IFR
      DO 270 JD=10,3000,10
      DECAY=EXP(-0.1*JD*LAMB)
      IFR(IJ,JD)=IFR(IJ,JD)*DECAY
      JZ=JD-10
      IF(JD.EQ.10) CFR(IJ,JD)=IFR(IJ,JD)
      IF(JD.GT.10) CFR(IJ,JD)=CFR(IJ,JZ)+IFR(IJ,JD)
```

Table 4.18, Continued

FORTRAN Code to Calculate the Incremental and Cumulative Fractional Releases From Unsolidified Waste and Resin in a Carbon Steel Container

```
  270 CONTINUE
C     CALCULATION OF ANNUAL RELEASES
      DO 300 K=10,3000,10
      KZ=K/10
      M=K-10
      IF(KZ.EQ.1) AR(IJ,KZ)=CFR(IJ,K)
      IF(KZ.GT.1) AR(IJ,KZ)=CFR(IJ,K)-CFR(IJ,M)
  300 CONTINUE
  400 CONTINUE
C     OUTPUT
      WRITE(6,500) (EAD(NZ),NZ=1,8)
  500 FORMAT(8A10)
      WRITE(6,510)
  510 FORMAT(20X,'CS',20X,'SR',20X,'CO',/,3X,
     1'TIME',3X,3(4X,'CFR',8X,'IFR',4X),/)
      DO 520 NO=10,3000,10
      NQ=NO/10
      WRITE(6,515) T(NO),(CFR(NU,NO),AR(NU,NQ),NU=1,3)
  515 FORMAT(1P,7G10.4)
  520 CONTINUE
      GO TO 1
 1000 CONTINUE
      STOP
      END
**
```

Table 4.19

Input Parameters for Release Modeling

Case	Container Surface Area (cm^2)	Container Volume (cm^3)	Initial Exposed Surface Area (cm^2)	Time to First Pit (yrs)	Life of Container (yrs)	Diffusivity cm^2/sec		
						Cs	Sr	Co
1	$2.0x10^4$	$2.1x10^5$	---	4	86	$2x10^{-8}$	$2\ x10^{-9}$	$4\ x10^{-13}$
2	$2.0x10^4$	$2.1x10^5$	$2.0x10^4$	4	86	$2x10^{-8}$	$2\ x10^{-9}$	$4\ x10^{-13}$
3	$1.4x10^5$	$4\ x10^6$	---	12	215	$2x10^{-8}$	$2\ x10^{-9}$	$4\ x10^{-13}$
4	$2.0x10^4$	$2.1x10^5$	---	0.8	11	$6x10^{-6}$	$3.3x10^{-6}$	$1.5x10^{-8}$
5	$2.0x10^4$	$2.1x10^5$	$2.0x10^4$	0.8	11	$6x10^{-6}$	$3.3x10^{-6}$	$1.5x10^{-8}$
6	$1.4x10^5$	$4\ x10^6$	---	1	28	$6x10^{-6}$	$3.3x10^{-6}$	$1.5x10^{-8}$
7	$2.0x10^4$	$2.1x10^5$	---	4	86	---	---	---
8	$2.0x10^4$	$2.1x10^5$	$2.0x10^4$	4	86	---	---	---
9	$1.4x10^5$	$4\ x10^6$	---	12	215	---	---	---

4.9 TRU Isotopes

Only four trenches (2, 7, 11, and 24) received significant amounts of TRU waste, and of these the Pu was concentrated in the first three (~1950 Ci) and Am-241 went mostly to Trench 24 (48 Ci of a total of 54 Ci) (see Tables 2.25-2.27). The Am was likely mostly in the form of oxide - Gamma Industries' contribution, for example, was in the chemical form of AmO_2. The 1.0 Ci shipment from Kay Ray, Am-Be sources, was probably metallic. The chemical form of the ANL Pu (150 Ci sent to Trench 2) is not known, but was probably oxide waste from examination of irradiated fuel. Any metallic Pu there might have been would have been oxidized on exposure to air. Thus, the ANL Pu waste is assumed to have been all oxide.

The Kerr McGee shipments contained hydroxide, or hydrous oxide, waste solidified in cement, contaminated equipment and gloves from glove box use, and contaminated plastic bags from bagging in and out of glove boxes. Most of this contamination was stated by Kerr McGee personnel to have been hydroxide and sintered oxide, but some of it was in the form of Pu(IV) nitrate. There is a possibility that some of the Pu from other shippers was also in the form of nitrate or other soluble salt when shipped.

Although the aqueous chemistry of Pu(IV) is quite complicated, in water of pH values expected in most locations in the trenches (measured values of samples from trenches and wells range from 5 to 12)[47] the soluble salts would be converted to hydrous PuO_2, which is highly insoluble. While it might be argued that the PuO_2 would simply form on contaminated gloves, equipment, etc., and remain part of the waste form, we have chosen to consider that once soluble Pu has dissolved, it has been released to the trench, even though insoluble hydrous PuO_2 forms rapidly thereafter. On the basis of the models presented in Section 4.3 and Appendix C, 15% of this release will occur in the first year from drums breached on emplacement, and in the fifth year from the remainder of the drums. The curie amounts of the releases from any of the wastes reported cannot be estimated with any precision since the proportion that is in the form of soluble salts is not known. Kerr McGee

personnel stated that the highest proportion of their waste was sintered oxide, the next highest was hydrous PuO_2, and the nitrate was lowest. From this description, it is likely that of the order of 10% of the Kerr McGee waste was in nitrate form, so that of the order of 60, 80, and 40 Ci would be released to Trenches 2, 7, and 11, respectively. This is made up of roughly 3, 4, and 2 Ci, respectively of α-emitting Pu-239,240, the remainder being β-emitting Pu-241 with a 13-year half-life.

The Kerr McGee cement waste forms would be subject to leaching once the drums containing them were breached. Leach rates for hydraulic cement waste forms containing Pu in incinerator ash have been deteremined at BNL.[46] The rates determined in a modified IAEA leach test using different groundwaters were around 10^{-8} g/cm^2-d, or 10^{-7} g Pu/L.[46] The latter figure is equivalent to roughly 10^{-9} moles/L. This value is somewhat lower than the solubility of crystalline PuO_2 at pH between 6 and 7 ($\sim 10^{-8}$ moles/L).[47] The leaching rate from cement is thus presumably lower than from PuO_2 itself by the same factor. Accordingly, the dissolution rate of PuO_2 would be expected to be of the order of 10^{-7} g/cm^2-d. This is lower than the 10^{-6} g/cm^2-d matrix dissolution rate of UO_2 used in Section 4.7.2, and taken from Reference 39. Thus, the release rates of PuO_2 from both the cement waste forms and from PuO_2 contamination waste from both Kerr McGee and ANL would be even lower than those of UO_2 discussed in Section 4.7.2. Amounts of Pu released from PuO_2 would then be insignificant compared to those released from nitrate waste.

The same argument will apply to AmO_2. It is known that all the Am waste of Gamma Industries was in the chemical form of AmO_2 (see Memo to File, August 25, 1983, "Am-241 Shipments From Gamma Industries to Trenches 11 and 24," reproduced in Appendix B). However, the form of the Am in the waste of the other shippers is not known. Thus, whatever proportion, if any, was in the form of soluble Am salts would have to be considered as being released to the trenches (Trench 2 and particularly Trench 24) on first contact with water.

4.10 Summary of Isotopic Releases to Individual Trenches

Information on release of C-14, I-129, and TRU isotopes is complete in the sections devoted to them (4.5, 4.6, and 4.9, respectively). For the remaining isotopes of concern, there were multiple cases and different distributions of activity for each case for any given trench. The various contributions to annual incremental and cumulative releases for each trench and for each isotope have been collected and summed to arrive at the values for total isotopic release to each trench. The results are presented in the following sections (4.10.1 and 4.10.2).

4.10.1 H-3 Releases

Calculation of IFRs, CFRs, annual incremental releases and annual cumulative releases was described in Section 4.4.10. The incremental and cumulative releases for a period of 90 years are given for each trench in Table D.2

of Appendix D. An abbreviated form of this table has been prepared by excerpting values for some representative times, and is presented as Table 4.20. The full results are presented in graphical form (computer generated graphics) in Figures 4.3 and 4.4.

Table 4.20

Annual Incremental and Cumulative Releases of H-3

TIME (YEARS)	TRENCH 1 INCREMENTAL RELEASE-H3 (CURIES)	TRENCH 1 CUMULATIVE RELEASE-H3 (CURIES)	TRENCH 2 INCREMENTAL RELEASE-H3 (CURIES)	TRENCH 2 CUMULATIVE RELEASE-H3 (CURIES)
1	8.9E+01	8.9E+01	1.5E+02	1.5E+02
2	4.9E+01	1.4E+02	5.2E+01	2.0E+02
3	3.2E+01	1.7E+02	3.5E+01	2.3E+02
4	5.	1.8E+02	8.	2.4E+02
5	5.	1.8E+02	7.	2.5E+02
10	3.	2.0E+02	4.	2.7E+02
20	1.	2.1E+02	2.	3.0E+02
40	.1	2.2E+02	.2	3.1E+02
60	1.6E-02	2.2E+02	3.7E-02	3.2E+02
90	7.2E-04	2.2E+02	3.3E-03	3.2E+02

TIME (YEARS)	TRENCH 7 INCREMENTAL RELEASE-H3 (CURIES)	TRENCH 7 CUMULATIVE RELEASE-H3 (CURIES)	TRENCH 11 INCREMENTAL RELEASE-H3 (CURIES)	TRENCH 11 CUMULATIVE RELEASE-H3 (CURIES)
1	1.8E+01	1.8E+01	1.1E+01	1.1E+01
2	5.	2.3E+01	6.	1.7E+01
3	3.	2.7E+01	4.	2.0E+01
4	.3	2.7E+01	.6	2.1E+01
5	.3	2.7E+01	.5	2.1E+01
10	.2	2.8E+01	.3	2.3E+01
20	6.7E-02	2.9E+01	.1	2.5E+01
40	8.6E-03	3.0E+01	1.5E-02	2.6E+01
60	1.0E-03	3.0E+01	1.7E-03	2.6E+01
90	2.7E-05	3.0E+01	4.7E-05	2.6E+01

Table 4.20, Continued

Annual Incremental and Cumulative Releases of H-3

TIME (YEARS)	TRENCH 14A INCREMENTAL RELEASE-H3 (CURIES)	TRENCH 14A CUMULATIVE RELEASE-H3 (CURIES)	TRENCH 23 INCREMENTAL RELEASE-H3 (CURIES)	TRENCH 23 CUMULATIVE RELEASE-H3 (CURIES)
1	4.1E+01	4.1E+01	1.5E+01	1.5E+01
2	2.1E+01	6.2E+01	8.	2.3E+01
3	1.6E+01	7.9E+01	7.	3.0E+01
4	9.	8.8E+01	6.	3.5E+01
5	9.	9.7E+01	5.	4.0E+01
10	5.	1.3E+02	3.	5.9E+01
20	2.	1.6E+02	1.	7.8E+01
40	.3	1.8E+02	.1	8.7E+01
60	4.2E-02	1.8E+02	1.8E-02	8.8E+01
90	3.5E-03	1.8E+02	8.1E-04	8.8E+01

TIME (YEARS)	TRENCH 24 INCREMENTAL RELEASE-H3 (CURIES)	TRENCH 24 CUMULATIVE RELEASE-H3 (CURIES)	TRENCH 25C INCREMENTAL RELEASE-H3 (CURIES)	TRENCH 25C CUMULATIVE RELEASE-H3 (CURIES)
1	2.3E+01	2.3E+01	1.2E+01	1.2E+01
2	1.2E+01	3.5E+01	4.	1.6E+01
3	1.1E+01	4.5E+01	4.	2.0E+01
4	1.0E+01	5.5E+01	3.	2.3E+01
5	9.	6.4E+01	3.	2.6E+01
10	5.	9.6E+01	2.	3.8E+01
20	2.	1.3E+02	.7	4.9E+01
40	.2	1.4E+02	8.7E-02	5.4E+01
60	3.2E-02	1.4E+02	1.1E-02	5.5E+01
90	1.4E-03	1.5E+02	4.3E-04	5.5E+01

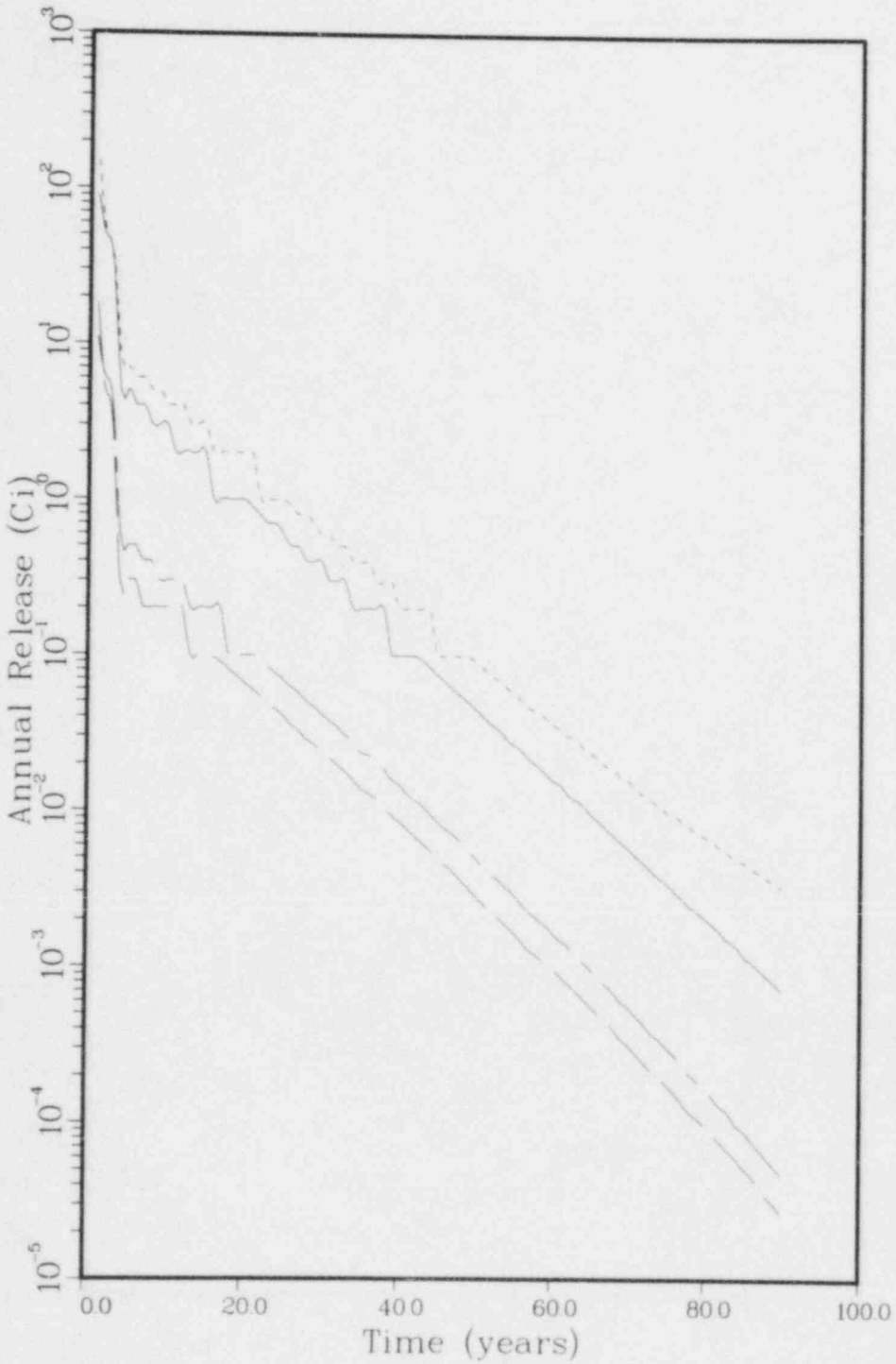

Figure 4.3. Annual releases from four trenches vs time after burial.
_____ Trench 1; --- Trench 2;
_____ . _____ Trench 7;
_____ - _____ Trench 11.

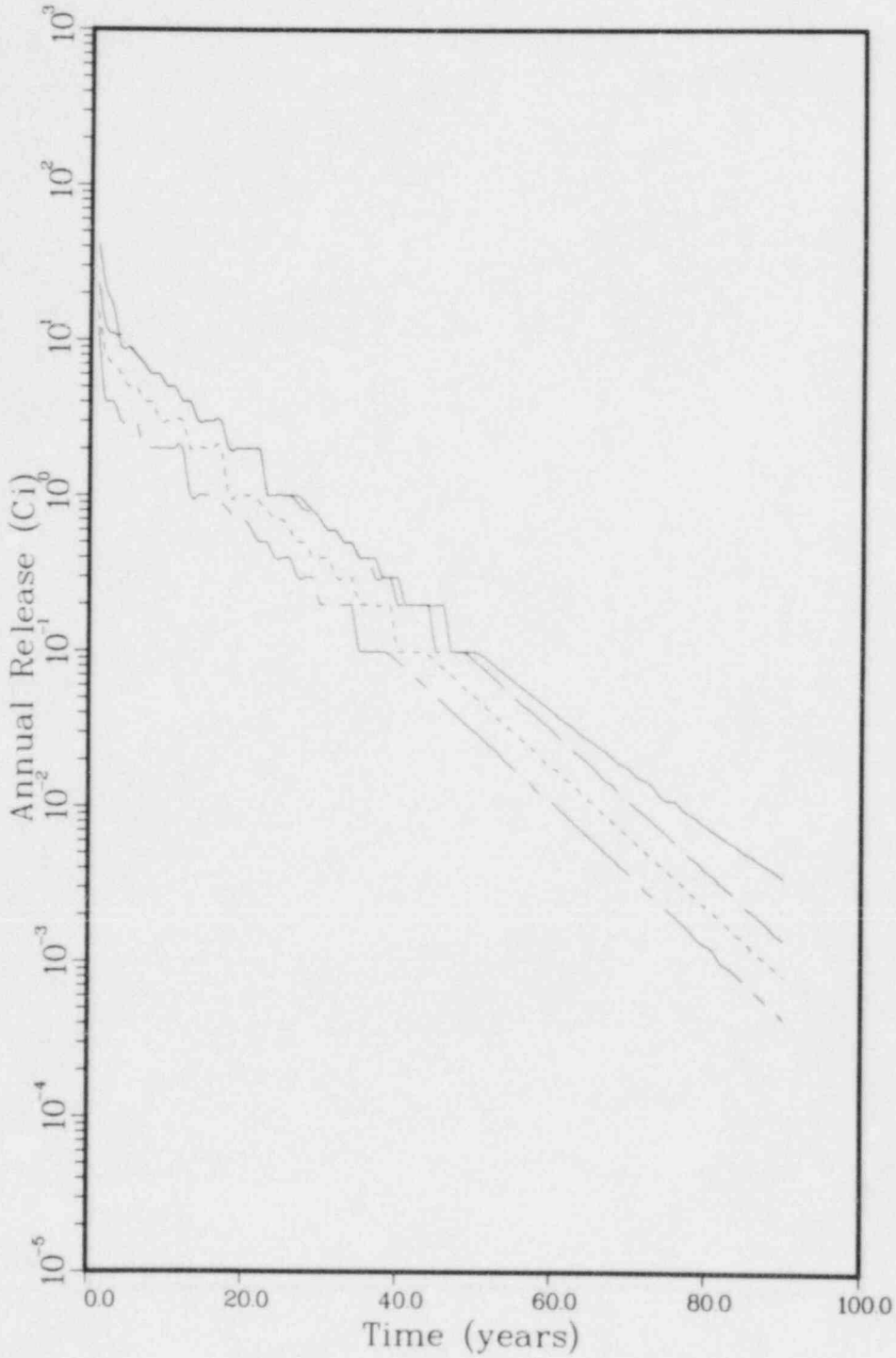

Figure 4.4. Annual releases from four trenches vs time after burial.
_____ Trench 14A; --- Trench 23;
_____ . _____ Trench 24;
_____ - _____ Trench 25C.

As discussed in Section 4.4.10, the choice made regarding time to failure of gaskets in the University of Wisconsin scrubbers has a significant effect on release to Trench 2 in early years. The results given in Table 4.20 assume the gaskets last indefinitely. Calculations were also done on the assumption that the gaskets failed in the first year. The results are given, alongside the previous Trench 2 results, in Table 4.21. Obviously first year release is affected strongly, but incremental releases for later years would not be much changed. Cumulative releases would, of course, be more than doubled because of the large fraction of the trench inventory in the scrubbers, which is all released in the first year in the second scenario. In reality, the gaskets would likely last for several years, and a peak in incremental release would occur after a few years, followed by incremental releases close to (and in between) those of the two extremes.

Estimated incremental and cumulative releases to Trench 2 are higher than those to any other trench, even without the University of Wisconsin scrubbers. Ninety year cumulative releases to Trench 1 are about 2/3 those to Trench 2, while those to other trenches range downward from Trench 14A with roughly 1/2 the Trench 2 values. Trench 14A had the highest inventory, and its relatively low cumulative releases are at least partly due to the slower releases from the BNL vaults, which contained well over half the trench inventory.

Table 4.21

Trench 2 H-3 Release Alternatives for
Different Treatments of University of Wisconsin Scrubbers

No Access to Water			Water Access in First Year		
	TRENCH 2			TRENCH 2	
TIME (YEARS)	INCREMENTAL RELEASE-H3 (CURIES)	CUMULATIVE RELEASE-H3 (CURIES)	TIME (YEARS)	INCREMENTAL RELEASE-H3 (CURIES)	CUMULATIVE RELEASE-H3 (CURIES)
1	1.5E+02	1.5E+02	1	3.4E+02	3.4E+02
2	5.2E+01	2.0E+02	2	5.2E+01	3.9E+02
3	3.5E+01	2.3E+02	3	3.5E+01	4.2E+02
4	8.	2.4E+02	4	7.	4.3E+02
5	7.	2.5E+02	10	4.	4.6E+02
10	4.	2.7E+02	20	2.	4.9E+02
20	2.	3.0E+02	40	.2	5.0E+02
40	.2	3.1E+02	60	2.3E-02	5.0E+02
60	3.7E-02	3.2E+02	90	6.9E-04	5.0E+02
90	3.3E-03	3.2E+02			

One of the more important observations to be made regarding the data presented in Table 4.20 and Figures 4.3 and 4.4 is that first year releases appear to have dominated the overall cumulative releases. The estimated first year release values range from roughly one-fifth of the total cumulative release for several trenches to more than half for one trench (Trench 7). These first year releases would have consisted mainly of gaseous tritium (T_2/HT) from targets, broken glass bulbs, luminous paint, etc. In only one trench (Trench 25C), is it estimated that liquid release (i.e., leaching) would have exceeded gaseous release in the first year. The estimates of first year gaseous and liquid releases are shown in Table 4.22. These are based on the inventories given in Table 4.10 for each trench and for the different release scenarios described by Cases 1 to 8, which are summarized in Section 4.4.10. After the first year, no liquid release has been assumed to occur until the fifth year. Even after that, as explained in Appendix C, release by leaching represents only a small fraction (approximately 3%) of the total tritium release from general trash and liquid waste solidified by NECO, the only categories of waste subject to liquid release aside from the University of Wisconsin scrubbers in Trench 2. The latter have been considered as a special case earlier in this section and in Section 4.4.2.2.

Table 4.22

Estimated H-3 Release to Trenches During First Year

Trench	Release (Ci)		
	Total	Leached	Gaseous
1	89	14	75
2	150	60	90
7	18	3	15
11	11	4	7
14A	41	11	30
23	15	6	9
24	23	10	13
25C	12	8	4

The assumption that essentially none of the types of packaging used at Sheffield provide an effective barrier to gaseous release is considered realistic. Thus, values given for estimated first-year gaseous release are felt to represent fairly closely what actually occurred. Assumptions affecting liquid release are conservative, however, and actual first-year liquid releases are likely to have been smaller than those given in Table 4.22.

In the first place, it has been assumed in the release scenarios that general trash releases all its tritium on first contact with water. This is undoubtedly conservative. Although most of Amersham/Searle's waste was water-soluble, other types of waste such as tritiated oil would have a very low

solubility in water. Furthermore, the assumption was made that all tritium was released from waste in fiberboard boxes during the first year, since these boxes provide no barrier to liquid water. While it is true that such boxes cannot be counted on to prevent water contact with the waste, the extent of tritium release would depend on the fraction of waste which is water soluble, and on the amount of water reaching the waste. This latter amount may be quite small near the bottoms of the trenches, since, at Sheffield, an estimated average of only 2.5 in./yr, out of an average rainfall of 35 in./yr, reaches the water table some 10 ft. below the floors of the 30-ft. deep trenches.[3]

Another conservative assumption is that all the tritium would have been rinsed out of the 15% of breached drums in the first year. This again depends on the fraction of water-soluble tritium waste, and also on the type of breaching and the orientation of the drums. Only those drums whose lids had been opened could have permitted significant release, not those which merely developed small holes due to corrosion of dented areas. Even those with opened lids would have permitted release of a significant fraction of activity only if they were in a suitable orientation, i.e., more nearly horizontal (to allow water to flow in and out) rather than nearly vertically upright or upside down.

In view of these considerations, the first year liquid releases given in Table 4.22 represent the extreme case. Realistically, the amounts shown would have been released over a period of several years, rather than all in the first year and none in years 2 to 4. However, it is not possible to make a reasonable estimate of actual annual liquid releases in the first few years, so we have chosen to present the conservative estimates in Tables 4.20 and 4.22. It should be kept in mind that none of the foregoing alters the overall picture of gaseous release dominating throughout the burial site history. In fact, the realistic assessment of liquid release probabilities shows that first year gaseous release would have predominated even more strongly than indicated by the figures in Table 4.22, since actual liquid releases would have been smaller than those shown.

Gaseous release at West Valley has been shown by Matuszek to be a major pathway for uncontrolled release of radioactivity from the trenches.[48,49] Tritium, mostly as CH_3T, was the isotope in by far the largest concentration in the samples taken. (Methane production at West Valley is probably due largely to biogenic processes.[50]) Conditions at West Valley are quite different from those at Sheffield, so the same conclusions cannot automatically be drawn for Sheffield. However, trench gas samples taken by the U.S. Geological Survey at Sheffield showed considerable tritium.[49] The chemical form of the tritium was not stated, and was probably not determined. In any case, gaseous tritium was reported to have been released to trenches at Sheffield. Considering the types of tritium waste and the distribution of curie amounts which have been determined in the present study, there seems to be no doubt that a large fraction of the tritium release to Sheffield trenches has been, and will continue to be, gaseous. The fraction escaping to the atmosphere will depend on the chemical form and on soil conditions.

Essentially all CH_3T will escape, but part of the T_2/HT component will be converted to T_2O/HTO, and part of the latter will presumably reach the groundwater, as discussed in Section 4.2.1.3.

Another important point which is indicated by the tables, and particularly well illustrated by the curves of Figures 4.3 and 4.4 is the continual fall-off of H-3 release with time. After the substantial releases of the first few years (largely gaseous), subsequent releases are estimated to be relatively small, and decreasing. (These smaller releases will also be predominantly gaseous, probably even to a greater extent than those of the early years.) No large releases, in comparison to the first few years, are thus expected to occur during the time remaining before the buried tritium has decayed to insignificant levels.

4.10.2 Sr-90, Cs-137, and Co-60 Releases

Calculation of releases for these isotopes was described in two sections, Section 4.7 for non-fuel cycle Sr-90 and Cs-137, and Section 4.8 for fuel cycle Sr-90, Cs-137, and Co-60. The results are given in Appendix D. The incremental and cumulative releases by trench for fuel cycle waste are given in Table D.4, and the same releases for non-fuel cycle waste to Trenches 1 and 2 are given in Table D.5. The total Sr-90, Cs-137, and Co-60 releases, by trench, are given in Table D.6. Abbreviated presentations of the non-fuel cycle data and of the total releases are given in Tables 4.23 and 4.24.

Comparing these two tables, it can be seen that releases of Cs-137 to Trenches 1 and 2 are dominated by the non-fuel cycle contribution after the 5-year drum corrosion induction period. After this period, the non-fuel cycle Sr-90 represents the total of the incremental Sr-90 releases to Trenches 1 and 2. The estimated cumulative releases of Sr-90 of >100 Ci to each trench in the first 10 years are considered to be rather high.

Releases of Co-60 are comparable to, but generally somewhat lower than, those of Cs-137 in the first few years. After less than 10 years, the Co-60 incremental releases are lower than those of Cs-137 for all trenches, and soon become insignificant due to the decay of Co-60 with its 5.3 year half-life. Cumulative releases of Co-60 are therefore much less than those of Cs-137, varying from a factor of 2 less (Trenches 7 and 25C) to an order of magnitude less (Trench 24).

Table 4.23

Annual Incremental and Cumulative Releases of
Non-Fuel Cycle Sr-90 and Cs-137 to Trenches 1 and 2

TRENCH 1

TIME (YEARS)	INCREMENTAL RELEASE-CS (CURIES)	CUMULATIVE RELEASE-CS (CURIES)	INCREMENTAL RELEASE-SR (CURIES)	CUMULATIVE RELEASE-SR (CURIES)
1	0.	0.	0.	0.
2	0.	0.	0.	0.
3	0.	0.	0.	0.
4	0.	0.	0.	0.
5	0.	0.	0.	0.
6	1.2E+02	1.2E+02	4.5E+01	4.5E+01
7	1.3E+02	2.5E+02	5.2E+01	9.7E+01
8	3.0E+01	2.8E+02	1.2E+01	1.1E+02
9	2.4E+01	3.0E+02	9.	1.2E+02
10	2.4E+01	3.3E+02	9.	1.3E+02
15	3.	3.4E+02	1.	1.3E+02
20	1.	3.5E+02	.5	1.4E+02
30	1.	3.7E+02	.4	1.4E+02
50	.2	3.8E+02	5.8E-02	1.5E+02
100	1.6E-03	3.9E+02	5.2E-04	1.5E+02
150	1.5E-02	3.9E+02	4.6E-03	1.5E+02
300	1.3E-04	3.9E+02	3.0E-05	1.5E+02

TRENCH 2

TIME (YEARS)	INCREMENTAL RELEASE-CS (CURIES)	CUMULATIVE RELEASE-CS (CURIES)	INCREMENTAL RELEASE-SR (CURIES)	CUMULATIVE RELEASE-SR (CURIES)
1	0.	0.	0.	0.
2	0.	0.	0.	0.
3	0.	0.	0.	0.
4	0.	0.	0.	0.
5	0.	0.	0.	0.
6	1.1E+02	1.1E+02	9.9E+01	9.9E+01
7	1.3E+02	2.4E+02	1.1E+02	2.1E+02
8	2.8E+01	2.6E+02	2.5E+01	2.4E+02
9	2.3E+01	2.9E+02	2.1E+01	2.6E+02
10	2.2E+01	3.1E+02	2.0E+01	2.8E+02
15	3.	3.3E+02	2.	2.9E+02
20	1.	3.3E+02	1.	3.0E+02
30	1.	3.5E+02	1.	3.2E+02
50	.2	3.6E+02	.1	3.3E+02
100	1.5E-03	3.6E+02	1.1E-03	3.3E+02
150	1.4E-02	3.6E+02	1.0E-02	3.3E+02
300	1.2E-04	3.7E+02	8.6E-05	3.3E+02

Table 4.24

Total Annual Incremental and Cumulative Releases of Cs-137, Sr-90, and Co-60

TRENCH 1

TIME (YEARS)	INCREMENTAL RELEASE-CS (CURIES)	CUMULATIVE RELEASE-CS (CURIES)	INCREMENTAL RELEASE-SR (CURIES)	CUMULATIVE RELEASE-SR (CURIES)	INCREMENTAL RELEASE-CO (CURIES)	CUMULATIVE RELEASE-CO (CURIES)
1	7.	7.	7.0E-02	7.0E-02	3.1E+01	3.1E+01
2	.2	8.	7.3E-04	7.1E-02	2.2E-03	3.1E+01
3	.2	8.	5.5E-04	7.1E-02	1.5E-03	3.1E+01
4	.1	8.	4.5E-04	7.2E-02	1.1E-03	3.1E+01
5	9.	1.7E+01	8.4E-02	.2	2.6E+01	5.7E+01
6	1.3E+02	1.4E+02	4.5E+01	4.5E+01	1.9E+01	7.6E+01
7	1.4E+02	2.8E+02	5.2E+01	9.7E+01	1.4E+01	9.0E+01
8	3.4E+01	3.1E+02	1.2E+01	1.1E+02	9.	1.0E+02
9	2.7E+01	3.4E+02	9.	1.2E+02	6.	1.1E+02
10	2.6E+01	3.7E+02	9.	1.3E+02	4.	1.1E+02
15	7.	4.0E+02	1.	1.3E+02	4.	1.3E+02
20	2.	4.2E+02	.5	1.4E+02	.4	1.4E+02
30	1.	4.5E+02	.4	1.4E+02	2.5E-03	1.4E+02
50	.3	4.6E+02	5.8E-02	1.5E+02	6.6E-06	1.4E+02
100	2.0E-02	4.7E+02	5.8E-04	1.5E+02	4.6E-09	1.4E+02
150	2.0E-02	4.7E+02	4.6E-03	1.5E+02	5.3E-12	1.4E+02
300	2.1E-04	4.7E+02	3.0E-05	1.5E+02	0.	1.4E+02

TRENCH 2

TIME (YEARS)	INCREMENTAL RELEASE-CS (CURIES)	CUMULATIVE RELEASE-CS (CURIES)	INCREMENTAL RELEASE-SR (CURIES)	CUMULATIVE RELEASE-SR (CURIES)	INCREMENTAL RELEASE-CO (CURIES)	CUMULATIVE RELEASE-CO (CURIES)
1	6.	6.	5.9E-02	5.9E-02	2.5E+01	2.5E+01
2	8.6E-02	6.	2.7E-04	5.9E-02	1.4E-03	2.5E+01
3	6.4E-02	6.	2.0E-04	6.0E-02	9.7E-04	2.5E+01
4	5.2E-02	6.	1.7E-04	6.0E-02	7.2E-04	2.5E+01
5	7.	1.3E+01	7.1E-02	.1	2.2E+01	4.7E+01
6	1.2E+02	1.3E+02	9.9E+01	9.9E+01	1.6E+01	6.3E+01
7	1.4E+02	2.6E+02	1.1E+02	2.1E+02	1.1E+01	7.5E+01
8	3.1E+01	2.9E+02	2.5E+01	2.4E+02	8.	8.2E+01
9	2.6E+01	3.2E+02	2.1E+01	2.6E+02	5.	8.7E+01
10	2.4E+01	3.4E+02	2.0E+01	2.8E+02	3.	9.1E+01
15	7.	3.8E+02	2.	2.9E+02	3.	1.1E+02
20	2.	3.9E+02	1.	3.0E+02	.4	1.2E+02
30	1.	4.1E+02	1.	3.2E+02	2.0E-03	1.2E+02
50	.2	4.2E+02	.1	3.3E+02	4.3E-06	1.2E+02
100	7.8E-03	4.2E+02	1.1E-03	3.3E+02	3.0E-09	1.2E+02
150	1.6E-02	4.2E+02	1.0E-02	3.3E+02	3.4E-12	1.2E+02
300	1.5E-04	4.3E+02	6.6E-05	3.3E+02	0.	1.2E+02

Table 4.24, Continued

Total Annual Incremental and Cumulative Releases of Cs-137, Sr-90, and Co-60

TRENCH 7

TIME (YEARS)	INCREMENTAL RELEASE-CS (CURIES)	CUMULATIVE RELEASE-CS (CURIES)	INCREMENTAL RELEASE-SR (CURIES)	CUMULATIVE RELEASE-SR (CURIES)	INCREMENTAL RELEASE-CO (CURIES)	CUMULATIVE RELEASE-CO (CURIES)
1	.6	.6	4.6E-03	4.6E-03	2.	2.
2	.2	.8	1.5E-03	6.1E-03	7.8E-03	2.
3	.5	1.	3.4E-03	9.5E-03	1.8E-02	2.
4	.4	2.	2.5E-03	1.2E-02	1.3E-02	2.
5	.9	2.	7.4E-03	1.9E-02	1.	3.
6	.8	3.	6.6E-03	2.6E-02	1.	4.
7	.7	4.	5.5E-03	3.2E-02	.8	5.
8	.6	5.	4.5E-03	3.6E-02	.5	6.
9	.5	5.	3.8E-03	4.0E-02	.3	6.
10	.4	6.	3.2E-03	4.3E-02	.2	6.
15	.5	8.	3.8E-03	6.2E-02	.2	8.
20	.3	1.0E+01	1.8E-03	7.4E-02	2.8E-02	8.
30	9.0E-02	1.1E+01	5.2E-04	8.5E-02	3.3E-04	8.
50	4.1E-02	1.2E+01	2.2E-04	9.2E-02	1.1E-05	8.
100	4.8E-03	1.3E+01	2.5E-05	9.7E-02	1.0E-08	8.
150	1.2E-03	1.4E+01	3.5E-06	9.7E-02	1.1E-11	8.
300	2.2E-05	1.4E+01	5.5E-08	9.7E-02	0.	8.

TRENCH 11

TIME (YEARS)	INCREMENTAL RELEASE-CS (CURIES)	CUMULATIVE RELEASE-CS (CURIES)	INCREMENTAL RELEASE-SR (CURIES)	CUMULATIVE RELEASE-SR (CURIES)	INCREMENTAL RELEASE-CO (CURIES)	CUMULATIVE RELEASE-CO (CURIES)
1	2.	2.	1.6E-02	1.6E-02	4.	4.
2	4.	6.	2.7E-02	4.3E-02	.1	5.
3	9.	1.5E+01	6.5E-02	.1	.4	5.
4	6.	2.1E+01	4.9E-02	.2	.2	5.
5	7.	2.9E+01	5.8E-02	.2	4.	9.
6	7.	3.6E+01	5.4E-02	.3	3.	1.2E+01
7	6.	4.2E+01	4.9E-02	.3	2.	1.4E+01
8	6.	4.8E+01	4.5E-02	.4	1.	1.5E+01
9	5.	5.3E+01	4.1E-02	.4	1.	1.6E+01
10	5.	5.8E+01	3.9E-02	.4	.6	1.7E+01
15	4.	8.1E+01	3.4E-02	.6	.6	2.0E+01
20	3.	9.9E+01	2.5E-02	.8	8.7E-02	2.1E+01
30	1.	1.2E+02	7.6E-03	.9	3.9E-03	2.2E+01
50	.3	1.3E+02	2.9E-03	1.	2.0E-04	2.2E+01
100	2.5E-02	1.4E+02	2.8E-04	1.	1.9E-07	2.2E+01
150	6.2E-03	1.4E+02	1.8E-05	1.	2.1E-10	2.2E+01
300	1.1E-04	1.4E+02	2.8E-07	1.	0.	2.2E+01

Table 4.24, Continued

Total Annual Incremental and Cumulative Releases of Cs-137, Sr-90, and Co-60

TRENCH 14A

TIME (YEARS)	INCREMENTAL RELEASE-CS (CURIES)	CUMULATIVE RELEASE-CS (CURIES)	INCREMENTAL RELEASE-SR (CURIES)	CUMULATIVE RELEASE-SR (CURIES)	INCREMENTAL RELEASE-CO (CURIES)	CUMULATIVE RELEASE-CO (CURIES)
1	9.	9.	4.9E-02	4.9E-02	1.3E+01	1.3E+01
2	4.	1.3E+01	2.4E-02	7.3E-02	.1	1.3E+01
3	7.	2.0E+01	4.9E-02	.1	.2	1.4E+01
4	6.	2.6E+01	3.7E-02	.2	.2	1.4E+01
5	1.1E+01	3.7E+01	8.0E-02	.2	1.2E+01	2.6E+01
6	1.3E+01	5.0E+01	8.0E-02	.3	9.	3.5E+01
7	1.0E+01	6.0E+01	6.6E-02	.4	6.	4.1E+01
8	9.	6.9E+01	5.6E-02	.4	4.	4.5E+01
9	8.	7.7E+01	4.9E-02	.5	3.	4.8E+01
10	7.	8.4E+01	4.3E-02	.5	2.	5.0E+01
15	7.	1.2E+02	4.5E-02	.8	2.	6.1E+01
20	5.	1.5E+02	2.6E-02	.9	.2	6.4E+01
30	2.	1.8E+02	1.0E-02	1.	3.8E-03	6.4E+01
50	1.	2.2E+02	5.0E-03	1.	1.6E-04	6.4E+01
100	.2	2.5E+02	6.4E-04	1.	1.4E-07	6.4E+01
150	4.2E-02	2.5E+02	1.2E-04	1.	1.6E-10	6.4E+01
300	7.7E-04	2.5E+02	1.9E-06	1.	0.	6.4E+01

TRENCH 23

TIME (YEARS)	INCREMENTAL RELEASE-CS (CURIES)	CUMULATIVE RELEASE-CS (CURIES)	INCREMENTAL RELEASE-SR (CURIES)	CUMULATIVE RELEASE-SR (CURIES)	INCREMENTAL RELEASE-CO (CURIES)	CUMULATIVE RELEASE-CO (CURIES)
1	9.	9.	5.6E-02	5.6E-02	1.8E+01	1.8E+01
2	5.	1.4E+01	2.7E-02	8.3E-02	.1	1.9E+01
3	9.	2.3E+01	6.2E-02	.1	.3	1.9E+01
4	7.	3.0E+01	4.7E-02	.2	.2	1.9E+01
5	1.3E+01	4.4E+01	.1	.3	1.6E+01	3.5E+01
6	1.5E+01	5.9E+01	9.7E-02	.4	1.2E+01	4.7E+01
7	1.2E+01	7.0E+01	8.0E-02	.5	8.	5.5E+01
8	1.0E+01	8.1E+01	6.9E-02	.5	6.	6.1E+01
9	9.	9.0E+01	6.0E-02	.6	4.	6.4E+01
10	8.	9.8E+01	5.2E-02	.7	2.	6.7E+01
15	8.	1.4E+02	5.6E-02	.9	2.	8.1E+01
20	5.	1.7E+02	3.2E-02	1.	.3	8.5E+01
30	2.	2.1E+02	1.1E-02	1.	5.1E-03	8.5E+01
50	1.	2.4E+02	5.2E-03	2.	2.1E-04	8.5E+01
100	.1	2.7E+02	6.3E-04	2.	1.9E-07	8.5E+01
150	3.6E-02	2.7E+02	1.0E-04	2.	2.1E-10	8.5E+01
300	6.6E-04	2.7E+02	1.6E-06	2.	0.	8.5E+01

Table 4.24, Continued

Total Annual Incremental and Cumulative Releases of Cs-137, Sr-90, and Co-60

TRENCH 24

TIME (YEARS)	INCREMENTAL RELEASE-CS (CURIES)	CUMULATIVE RELEASE-CS (CURIES)	INCREMENTAL RELEASE-SR (CURIES)	CUMULATIVE RELEASE-SR (CURIES)	INCREMENTAL RELEASE-CO (CURIES)	CUMULATIVE RELEASE-CO (CURIES)
1	8.	8.	5.2E-02	5.2E-02	9.	9.
2	9.	1.7E+01	6.0E-02	.1	.3	9.
3	1.9E+01	3.6E+01	.1	.3	.8	1.0E+01
4	1.4E+01	5.1E+01	.1	.4	.5	1.0E+01
5	1.7E+01	6.7E+01	.1	.5	8.	1.8E+01
6	1.8E+01	8.5E+01	.1	.6	6.	2.4E+01
7	1.5E+01	1.0E+02	.1	.7	4.	2.8E+01
8	1.4E+01	1.1E+02	9.9E-02	.8	3.	3.1E+01
9	1.3E+01	1.3E+02	9.2E-02	.9	2.	3.3E+01
10	1.2E+01	1.4E+02	8.6E-02	1.	1.	3.4E+01
15	1.0E+01	1.9E+02	7.4E-02	1.	1.	4.1E+01
20	8.	2.4E+02	5.5E-02	2.	.2	4.3E+01
30	3.	3.0E+02	1.9E-02	2.	8.5E-03	4.4E+01
50	1.	3.3E+02	7.6E-03	2.	4.3E-04	4.4E+01
100	.1	3.6E+02	8.1E-04	3.	4.1E-07	4.4E+01
150	3.1E-02	3.6E+02	9.0E-05	3.	4.5E-10	4.4E+01
300	5.8E-04	3.6E+02	1.4E-06	3.	0.	4.4E+01

TRENCH 25C

TIME (YEARS)	INCREMENTAL RELEASE-CS (CURIES)	CUMULATIVE RELEASE-CS (CURIES)	INCREMENTAL RELEASE-SR (CURIES)	CUMULATIVE RELEASE-SR (CURIES)	INCREMENTAL RELEASE-CO (CURIES)	CUMULATIVE RELEASE-CO (CURIES)
1	1.	1.	9.0E-03	9.0E-03	4.	4.
2	.3	2.	9.5E-04	9.9E-03	1.5E-03	4.
3	.2	2.	8.5E-04	1.1E-02	1.8E-03	4.
4	.2	2.	6.8E-04	1.1E-02	1.3E-03	4.
5	1.	3.	1.0E-02	2.2E-02	3.	6.
6	2.	5.	9.7E-03	3.1E-02	2.	8.
7	1.	6.	7.4E-03	3.9E-02	1.	1.0E+01
8	.9	7.	5.8E-03	4.5E-02	1.	1.1E+01
9	.8	8.	4.5E-03	4.9E-02	.7	1.2E+01
10	.6	8.	3.5E-03	5.3E-02	.4	1.2E+01
15	.8	1.2E+01	5.6E-03	7.9E-02	.4	1.4E+01
20	.4	1.5E+01	1.9E-03	9.4E-02	4.9E-02	1.5E+01
30	.2	1.8E+01	7.8E-04	.1	3.1E-04	1.5E+01
50	.1	2.2E+01	4.5E-04	.1	3.9E-06	1.5E+01
100	2.1E-02	2.5E+01	6.6E-05	.1	3.0E-09	1.5E+01
150	5.3E-03	2.6E+01	1.5E-05	.1	3.4E-12	1.5E+01
300	9.9E-05	2.6E+01	2.4E-07	.1	0.	1.5E+01

5. SUMMARY AND CONCLUSIONS

5.1 Trench Inventories

- An inventory has been made of the contents of Trenches 1, 2, 7, 11, 14A, 23, 24, and 25C at the Sheffield LLW burial site. For this purpose, microfilm copies of the radioactive shipment records (RSRs) were made available by the site operator (US Ecology, formerly NECO). Using the RSRs, compilation was made of the amounts of relevant isotopes with half-life >5 yrs shipped to each trench of concern.

- The compilation was done with the help of a data base set up on BNL's CDC 6600 computers using Intel Corporation's System 2000 data base management system. Information from some 1700 non-fuel cycle RSRs and 3200 fuel cycle RSRs was stored.

- On the basis of information supplied by nuclear power plant operators on isoto ic composition of their waste, estimates of the trench inventories of fuel cycle Cs-137 and Co-60 have been made. Total amounts sent to the eight trenches covered by this study were estimated at about 5900 and 6300 Ci, respectively. The fuel cycle Sr-90 inventory is taken to be 1% of the Cs-137.

- Tritium amounts in the fuel cycle waste were estimated to be very much smaller than those in the non-fuel cycle waste, with a total of about 40 Ci in the eight trenches.

- Estimated H-3 inventory in non-fuel cycle waste was about 2300 Ci in all eight trenches. Distribution among the trenches varied widely, and ranged from 35 Ci in Trench 7 to >700 Ci in Trench 14A. Type of waste and size of individual shipments also varied greatly in the different trenches.

- Inventories of C-14 and I-129 were estimated at about 100 Ci and 1 mCi, respectively, in all eight trenches. Only four trenches received I-129, in six shipments. Most shipments were a few μCi or less, while one contained essentially the whole 1 mCi inventory.

- Non-fuel cycle inventories of Cs-137 and Co-60 were small compared to fuel cycle contributions, except for Cs-137 in Trenches 1 and 2 where they were considerably larger. Non-fuel cycle Sr-90 inventories predominated in Trenches 1 and 2 and were comparable to the fuel cycle for the other trenches. Trenches 1 and 2 received a number of unusually large non-fuel cycle shipments of Sr-90 and Cs-137, totaling approximately 1300 and 2000 Ci, respectively, in the two trenches.

- Four trenches received insignificant amounts of TRU waste. However, rather large amounts were shipped to Trenches 2, 7, 11, and 24. These amounts are estimated at approximately 2000 Ci of Pu and 50 Ci of

Am-241. About 95% of the Pu activity was due to β-emitting Pu-241, the remainder to Pu-239,240. The Pu-241 will decay with a half-life of 14.7 years to 65 Ci of α-emitting Am-241. After late 1976, TRU waste was no longer accepted at Sheffield.

- A recurring theme is the wide divergence in the non-fuel cycle shipments to the various trenches, both in isotopic distribution and in waste category. This is due to several factors, such as changing research programs at institutions and changing business ventures of industrial firms. Fuel cycle waste is much more uniform in terms of isotopic composition and waste type, and variation in amounts among trenches is not generally great because of the large volumes shipped to all trenches.

- Previous estimates of isotopic site inventories were made by NUS Corporation[1] and by an interagency task force.[2] Based on results of the present study, the H-3 site inventory estimated by the task force is probably low by a factor of 2, while the NUS estimate is much lower still. The NUS estimate for C-14 site inventory may be somewhat high, but is considered to be within a factor of 2 of what is actually present. The Interagency Task Force estimate is much too low.

- The practices which led to the wide divergence in inventory estimates by NUS Corp. and the Interagency task force include reliance on general surveys, extrapolation from the experience at one trench to the entire site, and use of summaries of shipment information instead of examination of individual RSRs. Such methods have their place when an order of magnitude estimate is all that is required, or little time is available. However, it must be realized that the results they produce will be subject to large uncertainties.

5.2 Estimates of Isotopic Release Rates

- As was anticipated at the initiation of the study, the foundations for the predictions of radionuclide release to a disposal trench were extremely sparse. The modeling presented herein is highly speculative, but believed to be state-of-the-art. This area is a prime area for further research.

- In connection with the modeling done to estimate release of activity to the trenches, exact descriptions of waste forms and containers were often not available, and assumptions had to be made. These assumptions were made conservatively so as not to underestimate possible releases. At the same time, an attempt was made to keep the assumptions realistic.

- Estimates of isotopic release rates involved modeling the behavior of a number of different container-waste form combinations under the

conditions found in the trench environment. Both containers and waste forms of standard types, and those which are non-routine, have been considered.

- Information on waste form was rarely given on non-fuel cycle RSRs. This was obtained for shipments of relatively large amounts of activity by contacting the generators. These generators accounted for roughly 90% of the H-3 waste in most trenches. In certain cases, information on special containers was also required. These cases included BNL's concrete vaults, the 3M Co. lead-lined drums, and ANL's aluminum primary containers (MAP tubes).

- Fuel cycle waste information which was not always supplied on the RSRs and which was required for modeling included whether or not the waste was solidified, and the nature of the solidification agent. A detailed study was made of one trench (Trench 24) to obtain a breakdown into the amounts of the different isotopes (Co-60, Cs-137, and Sr-90), which were contained in unsolidified waste and in concrete and urea-formaldehyde resin, in either drums or liners. The proportions arrived at were applied to the other trenches.

- Releases from the fuel cycle waste were estimated by developing an expression for incremental fraction release for each of nine different package-waste form combinations, multiplying the resulting fractions by the isotope inventory applying to each case, and summing for each trench. Estimated Co-60 releases were comparable to those for Cs-137 in the early years, but became quite small due to the radioactive decay of the Co-60 ($t_{1/2}$ = 5.3 yr). Sr-90 cumulative releases were estimated to be of the order of a curie for most trenches, due to its small initial inventory.

- Release of H-3 was modeled using appropriate scenarios for the different types of waste involved. Eight cases were developed, for which incremental fraction releases (IFRs) were calculated. Then the H-3 inventories applying to each case for each trench were multiplied by the IFRs and summed to obtain the estimated total releases for each trench. Cumulative releases after 20 years ranged from 25 Ci (Trench 11) to 300 Ci (Trench 2).

- In general, H-3 releases were greater for trenches with higher inventories. In the case of Trench 14A, however, which had the highest inventory of all the trenches covered in this study, the cumulative releases were only half that of Trench 2, which had the next highest inventory. This was due principally to the slower releases from BNL vaults, which made up over half the trench inventory.

- The predominant mode of tritium release is gaseous rather than liquid. This is because most of the waste involved compounds (e.g., TiT targets, tritiated organic polymers in luminous paint, and other tritiated organics) which lose tritium as a gas and leach to a negligible extent with water. Tritium gas released to the trench may fairly

rapidly be converted to tritiated water by bacteria in the soil.[22] In dry periods this will percolate to the surface as vapor, but in wet periods some of it will be incorporated in groundwater.

- Estimates of C-14 fractional releases indicated that initial values, due to radiolysis alone, would be relatively small. However, releases would increase in later years, because of increasingly rapid leaching as drum corrosion progressed, until total release would occur, probably in less than 50 years.

- Non-fuel cycle Sr-90 and Cs-137 releases were estimated for the special lead-lined containers in Trenches 1 and 2. Since fuel cycle Sr-90 inventories were relatively small and the non-fuel cycle quite large for Trenches 1 and 2, the non-fuel cycle Sr-90 releases to these trenches constitute essentially the complete amounts released. Non-fuel cycle Cs-137 releases were considerably higher than fuel cycle, due to the soluble nature of the waste, but occurred in different time periods from those of the fuel cycle releases.

- I-129 was estimated to be completely released soon after burial (probably in the first year), due to the soluble nature of the waste form (NaI).

- TRU isotopes were mostly in the form of oxides, but it is estimated that roughly 10% of the Pu (~200 Ci) was in the form of nitrate. Release from the oxides is considered to be too slow to be significant. However, due to uncertainties concerning the solution chemistry of Pu(IV) nitrate, it can be considred that all the Pu nitrate waste would have been released (to Trench 2, 7, and 11) on first contact with water, the exact time depending on extent of drum breaching and rate of drum corrosion.

6. REFERENCES

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APPENDIX A

SNM AND SOURCE MATERIAL RSRs

List of RSRs for shipments of SNM and source material to Trenches 2 and 24 from which information was not entered in the BNL data base.

Trench 2

HILL 10/25/68	3336	3634	11564	5084
UNC 11/04/68	11573	11629	11570	5085
HILL 11/20/68	2272	3633	UNC 5/25/70	5095
HILL 12/03/68	11563	3621	4223	5105
HILL 12/12/68	UNC 8/25/69	3622	4224	5113,4
UNC 01/03/69	11547	11567	4371	4229
HILL 01/14/69	11628	3648	4226	4230
HILL 02/20/69	11575	11569	4221	4232
HILL 03/28/69	11574	11571	4228	10083
HILL 04/24/69	3643	11572	3927	10084

Trench 24

20017
9901
21070 (Kerr McGee 10/24/75)
20020
21302,3
21070 (Kerr McGee 11/11/75)
21070 (Kerr McGee 11/19/75)
21070 (Kerr McGee 11/25/75)
21070 (Kerr McGee 12/02/75)
21070 (Kerr McGee 12/29/75)
20910
21070 (Kerr McGee 12/29/75)
22705
KERR 03/30/76
20016

Explanatory Note:

In the first few years of operation at Sheffield, a great many RSRs did not have numbers (e.g., the first ten listed under Trench 2). For entry into the BNL data base, a shipment with no RSR number was identified by an abbreviated form of the shipper's name, followed by the shipment date. In the case of Trench 24, one shipper sent a number of shipments with the same RSR number (21070) - the only example encountered of this kind of occurrence. To permit identification of these particular RSRs, we have given the name of the shipper and the date of each shipment in the list above.

APPENDIX B

INFORMATION FROM GENERATOR CONTACTS

Memos to File containing information obtained from shippers regarding isotopic composition, waste form, and waste packaging, of significantly large shipments to the eight trenches included in this study.

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: March 21, 1983

TO: File

FROM: C. R. Kempf CRK

SUBJECT: Waste Form Information from Generators, Sheffield Site - Ames Laboratory, Tritium

Robert Staggs, the Radiation Safety Officer for Ames Laboratory (515-865-2153), was contacted on March 9, 1983 for information about the tritium waste shipped to Sheffield trenches 7, 11, and 23. The following information was obtained in a telephone conversation:

- The tritium waste consisted of neutron generator targets (obtained from Nuclear Chicago) and liquid scintillation vials.
- The targets were copper or aluminum backings on which thin coatings of titanium had been vacuum deposited and the titanium hydrided. The activity levels were typically 3-5 Ci/in.^{2} while whole target activities generally were approximately 1000 mCi.
- Targets were discarded because they were dirty and/or no longer useful in the generator. Prior to disposal they were bagged in 3 or 4 mil polyethylene and compacted
- Contents of liquid scintillation vials (LSV, nearly all plastic bottles) were emptied into glass beakers. The liquid was evaporated, leaving a residue. The empty bottles and the beakers containing the residues were then bagged, crushed, placed in drums and sent for disposal.
- Tritium wastes were frequently accompanied by low level dry lab refuse (wipes, rags, Kimwipes, etc.) sometimes contaminated with fission products.
- Wastes were shipped in 55-gallon mild steel drums with rubber gaskets and bolt ring seals.

A summary of the shipments, packages, contents, etc., follows:

Trench	Isotope, Amount, Form	RSR	Date
7	H-3, 13 Ci, targets	10375	4/10/74
11	H-3, 7 Ci total Item 21 - 6 Ci, targets Item 31 - 0.5 Ci, LSV Item 38 - 0.5 Ci, LSV	03314	4/30/75
23	H-3, 2.5 Ci total Drums 1, 2 - LSV Drum 28 - 1 Ci - target(s) Drum 36 - 1 Ci - target(s)	22159-60	10/26/76

CRK:kb
cc: R. E. Barletta
R. E. Davis
D. R. MacKenzie

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: March 22, 1983

TO: File

FROM: C. R. Kempff CRK

SUBJECT: Waste Form Information From Generators, Sheffield Site - Pathfinder Laboratory, C-14

Bill Perry, the Radiation Safety Officer for Pathfinder Laboratory (314-569-0681), was contacted by telephone on March 17, 1983 for information about the waste forms of several C-14 shipments sent to the Sheffield site. The wastes were all of the same type: C-14 labeled solid organics, mainly aromatics. Typical specific activities ranged from 5 mCi/mmol to 20 mCi/mmol. Frequently, lab trash in the form of paper, glass, and plastic was disposed of with the organics. All wastes were placed in 55-gallon mild steel drums with bolt ring seals.

The shipments referred to in this memo are given below:

Trench	RSR	Isotope and Amount	Date
14A	29541	C-14, 14 Ci	9/13/77
14A	36358	C-14, 6 Ci	12/19/77
14A	29306	C-14, 6 Ci	3/01/78
23	01558	C-14, <6 Ci	8/03/76

CRK:kb
cc: R. E. Barletta
R. E. Davis
D. R. MacKenzie
J. Smalley

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: March 24, 1983

TO: File

FROM: D. R. MacKenzie DRM

SUBJECT: H-3 Shipment of 6/23/76 (RSR 21758) to Trench 25C by University of Missouri (Rolla)

According to the RSR, the above shipment contained 8.2 Ci of H-3 in 150 gallons of liquid waste. In discussion with Mr. Ray Bono (telephone conversation, March 21, 1983), these figures were confirmed. He suggested talking with Mr. Tom Froelich, now with Washington Power and Light, who was the person who prepared the waste for shipment, completed the forms, and sent the shipment.

Mr. Froelich remembered the shipment well, including the method of packaging in 5-gal cans which were packed in absorbent inside standard DOT-approved 55-gal steel drums. He also remembered talking with NECO personnel at Sheffield who described to him the operation of their solidification unit and assured him he could ship the waste as liquid and they would solidify it, even though it consisted largely of toluene. Mr. Froelich also stated that the amount of C-14 in the waste was small compared to that of H-3.

DRM/nmy
cc: R. E. Barletta
R. E. Davis
C. R. Kempf
J. Smalley

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: March 25, 1983

TO: File

FROM: D. R. MacKenzie DRM

SUBJECT: C-14 in Shipment of 5/02/74 (RSR 2531) by Michigan State University

The above shipment to Trench 7 had three packages listed on the RSR as containing 1 Ci of C-14 each. University personnel contacted (telephone call to Warren Malchman, March 23, 1983) stated that no such amounts of C-14 ever existed at the University. Since the RSR had been filled in to indicate the large amounts, Mr. Malchman assumed someone had made an error in completing the form. He did some checking of University records and reported (telephone call, March 24, 1983) that the particular shipment and RSR had originated at an off-campus extension laboratory, and his office had not seen it before it was shipped. He felt that the person completing the RSR was unfamiliar with the forms, and was also unused to thinking or dealing in curie amounts, so had listed the number of millicuries in a column labelled "curies." Mr. Malchman had found purchase orders showing that the amounts of C-14 purchased for the extension laboratory (a few mCi) corresponded to the amounts disposed of after their experiments were completed. Our data base is thus in error and will be corrected accordingly.

DRM/nmy
cc: R. E. Barletta
R. E. Davis
C. R. Kempf
J. Smalley

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: March 28, 1983

TO: File

FROM: D. R. MacKenzie

SUBJECT: University of Minnesota Shipment of May 28, 1975; Sheffield RSR 1716

Mr. Jerry Steiger of the Environmental Health Department's Radiation Protection Division at the University of Minnesota was asked (telephone conversation of March 22, 1983) about the waste form and packaging of the H-3 and C-14 wastes reported in the above shipment. The amounts listed totalled 12.9 Ci of H-3 and 2.8 Ci of C-14. Mr. Steiger was certain there was an error in the RSR information since his recollection of shipments made in that period (indeed throughout the whole period in which the University used radioactive material) was that they were of the order of millicuries rather than curies.

In a telephone conversation on March 28, 1983, Mr. Steiger confirmed that the amounts listed on the RSR should indeed have been millicuries instead of curies. The column for amount of radioactivity on the RSR was labelled "curies", and in other RSRs, the personnel filling in the forms had added "milli" by hand, but had neglected to do so on this particular RSR.

Since the amounts in the shipment were only a few mCi, the matter of waste form and packaging was inconsequential and was not pursued.

DRM/nmy
cc: R. E. Barletta
R. E. Davis
C. R. Kempf
J. Smalley

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: March 28, 1983

TO: File

FROM: D. R. MacKenzie DRM

SUBJECT: Shipments by Midwest Research Institute Containing H-3 and C-14 - Amounts, Form and Packaging

Amounts of H-3 and C-14 obtained from our data base for two Midwest Research Institute shipments are summarized in the following table.

Table 1

Amounts of H-3 and C-14 Given in Data Base

Trench	RSR	Date	Packages	Isotopes Present	Amounts (mCi)
25C	21201	6/17/76	1-5	C-14	1 each
			6	H-3	1000
			7	H-3 and C-14	6000
23	32482	8/09/76	1,2	H-3 and C-14	2000 each
			3-7	C-14	100 each

In the shipment of August 9, 1976 (RSR 32482), a total of 0.5 Ci C-14 was apparently present (100 mCi in each of five packages) and it was of interest to know how much more was present in the 4 Ci of H-3 and C-14 in packages 1 and 2. In a telephone conversation (March 28, 1983) with Mr. Ed Ogilvie, the Radiation Safety Officer at Midwest Research Institute, it was determined that there was almost certainly only a few mCi of C-14 in the package. His records also showed that packages 3-7 contained only 100 µCi each rather than 100 mCi, so that our data base is in error. This is certainly more in line with the amounts for packages 1-5 in the shipment of June 17, 1976 (RSR 21201). He stated that the amount of C-14 in package 7 of that shipment was also almost certainly only a few mCi since that was the normal amount used in their experiments.

The H-3 waste (4 Ci and 7 Ci, respectively) for the two shipments was mostly contaminated lab ware (glass, metal, plastic, paper) with relatively small amounts of liquid waste sorbed by Vermiculite or other sorbent and placed in separate containers. The lab ware, sorbent containers, etc., were packed by individual laboratories in polyethylene bags and these were loaded by the radiation safety office into polyethylene liners of 3-4 mil thickness in standard 55-gal steel drums.

DRM/nmy
cc: R. E. Barletta
R. E. Davis
C. R. Kempf
J. Smalley

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: March 29, 1983

TO: File

FROM: C. R. Kempf CRK

SUBJECT: Waste Form Information From Generators, Sheffield Site - University of Wisconsin, Tritium and C-14

John Lorenz, the Radiation Safety Officer for the University of Wisconsin [(608) 262-8769], was contacted by telephone on March 25, 1983 for information about the tritium and carbon-14 waste shipped to Sheffield trenches 25C, 23, and 14A.

- Tritium wastes were composed mostly of four types of materials: tritium from liquid scintillation vial cocktails, labeled organic and aqueous materials (including significant quantities of tritiated water), tritium (and sodium-22) contaminated dog blood and brain cells, and contaminated lab refuse.

- Carbon-14 wastes consisted mainly of labeled compounds and contaminated lab refuse. The amounts of carbon-14 were consistently smaller than amounts of tritium by about two orders of magnitude.

- Proportions of organic to aqueous materials could not be specifically given. It was known that bicarbonate, cyanide and azide were among the labeled compounds.

- Several shipments contained liquid wastes. These materials were shipped in plastic and glass bottles, cushioned with vermiculite and placed in 30-gal steel drums. These drums (cushioned with vermiculite) were then placed in 55-gal mild steel drums which were all ring-bolt sealed.

- In one shipment (RSRs 12444,5, 5/27/76) solids were shipped in 4-1/2 cubic-ft. cardboard boxes.

A summary of the shipment information obtained appears in the following Table.

Summary of
University of Wisconsin Tritium and Carbon-14 Wastes in
Trenches 25C, 23, and 14A

Trench	Isotope, Amount, Form	RSR	Date
	Shipment 1		
25C	H-3, <12.9 Ci	12444,5	5/27/76
34 containers:	10 liquid 24 solid		
10 liquids:	55 gal with 30-gal drums LSV, labeled compounds, tritiated water Container amounts as listed (mCi): 79, 32, 138, 152, 38, 133, 141, 15.4, 81, and 103.		
24 solids:	Cardboard boxes (4-1/2 cu ft.) balance of activity of shipment: lab refuse paper, plastic gloves, some glass		
	Shipment 2		
23	H-3, <5.3 Ci C-14, <50 mCi	12447	10/28/76

(Most of H-3 in this shipment was from a H-3, Na-22 double-label experiment: dog blood and brain cells)

Some particular liquid-containing packages:
(ring-bolt sealed 55-gal drums with inner 30-gal drums)

(1) 50 mCi; H-3, S-35, Ca-45 general labeled liquids.

(2) 115 mCi; Zn-65, S-35, H-3, and Na-22 (from dog experiment, also some LSV)

(3) 85.5 mCi; unidentified mixture (some H-3)

(4) 607.9 mCi; unidentified origin, labeled compounds mixture of H-3, C-14, I-125, Cr-51.

(5) 1300 mCi; unknown origin (some H-3)

One solid package: (1) 3.1 Ci; mostly tritium contaminated lab refuse

Table, Continued

Summary of
University of Wisconsin Tritium and Carbon-14 Wastes in
Trenches 25C, 23, and 14A

Trench	Isotope, Amount, Form	RSR	Date
	Shipment 3		
14A	H-3, ~20 Ci total	36729,30	10/13/77

Liquid wastes: (55-gal with inner 30-gal drums)
total of 11 packages, 594 mCi,
LSV and aqueous

Solid wastes: (55-gal, ring-bolt sealed drums)

(1) 3400 mCi, paper trash contaminated with tritiated water.

(2) 772 mCi, paper trash contaminated with tritiated water.

(3) 6844 mCi, H-3, Cs-137, S-35, most of activity from H-3.

(4) 6302 mCi, mixture H-3, Cs-137, Cl-36, Na-22.

(5) 3085 mCi, mixture, similar to (4).

Packages (3), (4), and (5) mostly lab trash contaminated with labeled materials.

CRK/nmy
cc: R. E. Barletta
R. E. Davis
D. R. MacKenzie
J. Smalley

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: March 30, 1983

TO: File

FROM: C. R. Kempf CRK

SUBJECT: Waste Form Information From Generators, Sheffield Site - Dow Chemical, Tritium and Carbon-14

Rich Olson of Dow Chemical, Midland, Michigan, was contacted by telephone [(517) 636-5641] on March 29, 1983 for information about the tritium and carbon-14 waste shipped to Sheffield trenches 11 and 14A. (Tracy Parsons, who was unavailable, is the Radiation Safety Officer for Dow, [(517) 636-3205].)

Tritium and carbon-14 wastes in these shipments consisted of sealed sources, electron capture detector foils, animal carcasses, labeled organics (some LSV), contaminated soil and contaminated lab refuse.

One shipment (RSR 2282, 5/1/75, to trench 11) was made up of 42 drums, 40 of which contained solid materials and two of which contained liquid wastes. Drums were lined with thin (3-4 mil polyethylene) bags. Most of the 40 solid-containing drums were fiber-pack (cardboard/paper matrix) composition, while the two liquid-containing drums were steel. Liquids were held in gallon polyethylene bottles inside the 55-gal drums. Fiber-packs were closed by a clamp, while the steel drums were closed with a ring-bolt seal. The bulk of the waste volume consisted of animal carcasses; most of the H-3 activity was present as a target and a sealed source.

The other shipment (RSR 22527, 8/16/77, to trench 14A) consisted of 140 drums (with polyethylene liners) 37 of these have been accounted for as follows: 16 drums contained animal carcasses and lab plastic and paper waste in which H-3 and C-14 amounts were 0.128 mCi and 0.029 mCi, respectively (totals 2.048 Ci H-3 and 0.464 Ci, C-14); 21 drums each of which contained 0.5 mCi of C-14 contaminated soil. In addition, several specific high-activity items which were disposed of in a number of other drums are listed in the Table below. The remainder of the drums contained a variety of lab refuse and contaminated materials in which activities were much less significant.

In summary, the information obtained is given in the following Table.

Summary of
Dow Tritium and Carbon-14 Wastes in Trenches 11 and 14A

Trench	Isotope, Amount, and Form	RSR	Date
	Shipment 1		
11	H-3, 8 Ci	2282	5/01/75
	42 drums, bulk of volume - animal carcasses (1) H-3, 7 Ci, target (1) H-3, 1 Ci, sealed source C-14, 25-30 mCi, most C-14 present as labeled organics, few LSVs		
	Shipment 2		
14A	H-3 > 6.7 Ci C-14 >28.8 mCi 140 drums total	22527	8/16/77
	(16) drums - animal carcasses, 0.128 mCi H-3 and .029 mCi C-14 each (21) drums - C-14 contaminated soil, 0.5 mCi each (2) H-3, 500 mCi, EC detector (1) H-3, 3.2 Ci, solid source (1) H-3, 2500 mCi, sealed source (1) C-14, 12 mCi, solid (source?) (1) C-14, 5.8 mCi, lab refuse, labeled compounds		

CRK/nmy
cc: R. E. Barletta
R. E. Davis
D. R. MacKenzie
J. Smalley

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: March 30, 1983

TO: File

FROM: D. R. MacKenzie

SUBJECT: H-3 and C-14 in Shipments by Amersham/Searle Corporation and Amersham Corporation

Although Amersham/Searle Corporation and Amersham Corporation (name change in 1977) made no shipments containing strikingly large amounts of H-3 or C-14, the total amounts shipped to the trenches covered by our SOW placed them among the top five shippers. Also they sent shipments >1 Ci to all trenches except Trench 11. Details of the shipments are given in Table 1.

Table 1

Shipments >1 Ci of H-3 and C-14 sent by Amersham/Searle Corporation and Amersham Corporation

Trench	RSR	Date	Amount (Ci)[a]
7	11913	5/10/74	2.0
25C	12462	5/28/76	5.5
25C	12463	6/16/76	2.1
23	31689	9/09/76	3.5
23	31692	10/22/76	3.3
23	31693	12/03/76	2.7
14A	23334	12/12/77	9.1
14A	23335	2/03/78	1.6
14A	51143	3/31/78	2.6

Several other isotopes were normally present in packages with H-3 and/or C-14, particularly I-125, and frequently S-35 and Se-75, none of which are required for our inventory. The chemical form of almost all the waste was given as "water-based radiochemicals". Discussion with Linda Bagby at Amersham Corporation (telephone conversations, March 9, and March 31, 1983) elicited the following information:

1. Isotopes other than H-3 and C-14 were present in amounts considerably smaller than those of the H-3 and C-14. I-125, for example, would contribute a maximum of 100 mCi to the activity of any package, and usually would be much less than that. Amounts of S-35 and Se-75 were still less.

2. The curie amount of H-3 in the waste was generally about five times that of the C-14.

3. The "water-based radiochemicals" were organic compounds, most of them soluble in water or alcohol.

4. All of the waste was described as solid. Most of it would be in dry form, contaminating laboratory equipment such as vials and pipettes. However, some of it was in small amounts of liquid contained in vials, which were packed in absorbent.

5. The contaminated lab ware was placed in secondary plastic (acetate) containers, which were placed in polyethylene bags. The bags were packed 15-20 to a standard DOT 55-gal drum.

DRM/nmy
cc: R. E. Barletta
R. E. Davis
C. R. Kempf
J. Smalley

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: April 1, 1983

TO: File

FROM: J. F. Smalley J. F. S.

SUBJECT: Shipment Sent to Trench 14A of the Sheffield Burial Site by the Illinois Institute of Technology (IIT) on February 15, 1978 (Shipment Number IIT 2/15/78/ in the Data Base)

This shipment contains a large amount (74.3 curies) of tritium. Mr. Dave Derenzo (Radiation Safety Officer for IIT) was contacted and asked about the chemical form and packaging of the tritium waste. After consulting with other people at IIT, Mr. Derenzo said that, to the best of their recollection, the waste was unused tritium paint in its original metal containers. He also said that these containers of paint were probably packed in 55-gal drums. This may be contradictory because the RSR states that the entire shipment (which includes several radionuclides besides tritium) was buried in a single large (200 cu ft.) container. However, the 55-gal drums could have been packaged in this large container.

Since Hittman Nuclear Development Corporation packaged this shipment, they were contacted. Mr. Francis Dewberry of Hittman stated that, although he could not say for sure, this container was probably one of their steel liners. He is going to check with some other people and call back. However, from the information on the RSR (e.g., the weight of the entire shipment was only 5117 lbs) it seems likely that the container was made out of metal rather than concrete.

JFS/nmy
cc: R. E. Barletta
R. E. Davis
C. R. Kempf
D. R. MacKenzie

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: April 11, 1983

TO: File

FROM: C. Ruth Kempf CRK

SUBJECT: Waste Form Information from Generators, Sheffield Site
Mallinckrodt, Inc., Tritium

Roy Brown of Mallinckrodt Nuclear called on March 31, 1983 to provide the information requested in our letter to him of March 21, 1983. The letter asked for information on a Mallinckrodt shipment of 9.6 Ci of H-3 dated March 4, 1974. He said he had not been able to locate a file on that particular shipment but that he did have information on several other H-3 shipments which occurred around the same time. All of those shipments contained H-3 as tritiated water in small (~10ml) glass vials closed with rubber stoppers and metal crimp caps. These vials were packed in fiberboard drums 5.4 cu ft., 30" high, 12" diameter with polyethylene liners. The drums had metal lids and spring type latches with security wires.

On one or two occasions the liquids were shipped in small fiberboard/ cardboard boxes instead of the fiberboard drums.

CRK:cv
cc: R. E. Barletta
D. R. MacKenzie
J. F. Smalley

TIMEX
CORPORATION

BUILDING 19 ADAMS FIELD
POST OFFICE BOX 1676
LITTLE ROCK ARKANSAS 72203

April 21, 1983

Donald R. MacKenzie
Brookhaven National Laboratory
Associated Universities, Inc.
Upton, Long Island, New York 11973

Dear Mr. MacKenzie:

This letter is to answer the questions posed in your letter of April 6, 1983.

The shipments of radioactive waste from Timex Corporation in Little Rock, Arkansas resulted from the operation of applying tritium containing luminous paint to hands and dials for use in Timex watches. An NRC license allowing the operation was awarded after the conditions of 10 CFR 32.22 were satisfied. All the waste to which you refer meet the rigid standards listed in 10 CFR 32.22.

In order to respond to your specific questions, a brief description of the process is required. The luminous paint is purchased from a supplier with tritium only in the form of tritiated polystyrene. The tritiated polystrene, mixed thoroughly with a flourescent material, is supplied in ten gram vials with a specific activity ranging from 100 to 200 millicuries per gram. This powder is mixed with a polymeric paint that cures to a stable, inert luminous material that adheres strongly to the hands and dials.

The entirity of the radioactivity in the waste resulted from the tritium contained in the above described paint. The tritium waste consisted of luminous paint on scrap hands and dials, paper tissue used in cleaning, gloves, shoe covers and glass shipping vials.

The tritium waste was placed in a container obtained from NECO and was a standard DOT 55-gallon steel drum with a heavy plastic liner that could be tightly closed at the top. The loose waste was sealed in the plastic liner prior to sealing the top on the DOT drum.

continued...

Our Health Physics consultant is Dr. Daniel Mathews, professor, University of Arkansas. He quotes a half-life for release of tritium from the paint as in the range of 12 to 18 years and the principle tritium containing gases released in the molecular form of H_2, H_2O, CH_4, and C_2H_6.

I am not certain what "specific activity" means in this waste, but the specific activity of the initially prepared paint is in the range of 30 to 100 millicuries per gram, dependant on the specific paint and the method of mixing.

Yours very truly,

TIMEX CORPORATION

Daniel H. Woods
Human Resources Manager

DHW/bp

cc: Fazio

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: June 16, 1983

TO: File

FROM: D. R. MacKenzie

SUBJECT: Waste Form and Packaging in Large Shipments of Cs-137 and Sr-90 From 3M Co. to Sheffield Trenches 1 and 2.

Four 3M Co. shipments to Trench 1 and five to Trench 2 contained >100 Ci of Cs-137 and Sr-90. These are listed in Table 1. Sixteen other shipments of between 10 and 100 Ci were recorded, with a total of 1833 Ci Cs-137 and 1163 Ci Sr-90. The RSRs gave no information on form or packaging of the waste, except for the volumes of the packages and the fact that the waste was solid.

Table 1.

Cs-137 and Sr-90 Shipments >100 Ci From 3M Co. to Trenches 1 and 2

Trench	Date	Amount (Ci) Cs-137	Sr-90
1	5/02/67	395	250
1	6/11/68	153	
1	8/06/68	66.2	50
1	8/15/68	102.5	
2	11/26/68	118	150
2	1/21/69	219.5	
2	4/02/70	54.9	65.3
2	7/30/70	109.5	51.0
2	10/08/70	76	337

In a telephone conversation of June 1, 1983, Mr. M. Cobian of 3M Co. described the processes used in making sources, and the types of waste generated, in the period during which Trenches 1 and 2 were in operation.

A hot cell was used for handling the high curie levels of isotopes obtained from ORNL and incorporating them into "microspheres" (ceramic in the case of Cs-137 and Sr-90). The microspheres were transferred as needed to production facilities for fabrication into sources for use in the medical field. The operations involved at that stage consisted principally of loading the microspheres into metal tubes or needles in the amounts necessary to give the required dose rates for particular applications.

Mr. Cobian stated that a reasonable estimate of the chemical form of the Cs-137 and Sr-90 waste from the hot cell was one third chloride, one third nitrate and one third ceramic (from microspheres that were damaged, of the wrong size, or otherwise not meeting specifications). High activity waste from the hot cell included heavily contaminated equipment such as muffle furnaces, tongs and various containers. Waste from the production facilities was mostly faulty tubes and needles, e.g., damaged, leaking, or containing incorrect amounts. General low activity waste was generated by both the hot cell and production operations.

High activity waste from the hot cell was shipped in lead-lined 55 gallon drums. These drums were lined with 2 or 3 inches of lead, with the lids simply set on and sealed in the normal way with ring and gasket. No seal was made between the lead on the lid and that lining the body, for example by melting. The high activity waste from the production facilities, consisting mostly of tubes and needles, was gradually collected in lead buckets. When these were considered full enough, they were placed in concrete-lined 30-gallon or 55-gallon drums. As with the lead-lined drums, the concrete lids were simply placed in position and the metal lids sealed in the normal way. Thus, neither the lead nor the concrete linings provided a seal, but were merely required for shielding. Low activity wastes were normally shipped in fiberboard boxes.

DRM:cv
cc: Robert E. Barletta
Richard E. Davis
C. Ruth Kempf
John Smalley

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: August 8, 1983

TO: File

FROM: D. R. MacKenzie DRM

SUBJECT: I-129 Shipment to Trench 2

The BNL data base showed two shipments of I-129 to Trench 2. One, from Michigan State University, was listed as containing a package with 2 mCi of a mixture of H-3, C-14 and I-129. The other, from the University of Illinois, had a package with 0.2 mCi Co-57 and 1.0 mCi of I-129.

On examining the Michigan State University RSR, it was found impossible to distinguish the third digit in the isotopic mass number of the iodine for the package in question, but several other packages contained I-125. Health Physics personnel at MSU were contacted (telephone conversations with Warren Malchman of August 2 and 4), and their records showed that the isotope involved was I-125, not I-129. The BNL data base will be corrected accordingly.

Mr. Lorion Sanders of the Environmental Safety Department at the University of Illinois was asked about their package listed as containing I-129 (package six of shipment on 9/17/69, RSR Number 11422). In telephone conversations of August 3 and 5, Mr. Sanders confirmed that the material was, indeed, I-129. The amount listed was that given by the researcher, and was felt to be reasonably accurate. The isotope was <u>not</u> in a calibrated source but had been purchased for use in high energy physics experiments. It is probable that the waste consisted of unused material and some used targets, both probably having the chemical form of inorganic iodide. The professor who used and disposed of the isotope has retired. The container was a standard 55 gallon drum and packaging would have been in the usual polyethylene bags.

DRM:cv
cc: R. E. Barletta
R. E. Davis
C. R. Kempf
J. Smalley

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: August 11, 1983

TO: File

FROM: D. R. MacKenzie

SUBJECT: Large H-3 Shipments From University of Wisconsin to Trenches 1 and 2

The University of Wisconsin shipped 126 Ci of H-3 to Trench 1, and over 290 Ci to Trench 2, making it the largest single contributor to these trenches. The bulk of the ctivity - 121 Ci in Trench 1 and 269 Ci in Trench 2 - was contained in the six shipments listed in the Table, with amounts and forms as given on the RSRs.

Table

Large H-3 Shipments From
University of Wisconsin to Trenches 1 and 2

Trench	RSR	Date	Amount (Ci)	Form
1	-----	3/08/68	121	solid
2	11212	3/20/69	83	solid
2	11211	5/22/69	124.5	solid
2	3532	1/21/70	16.1	solid
2	-----	10/08/70	17.1	solid
2	3534	2/23/71	28.0	solid

Information regarding the waste packages in these shipments was obtained in telephone calls to John Lorenz, the University's Radiation Safety Officer, on May 27 and August 10.

NECO had recorded the activity in the March 8, 1968 shipment to Trench 1 as 11 Ci, but in our May 27 call, Mr. Lorenz confirmed that 121 Ci was, indeed, correct. It consisted of 12 packages each containing 10 Ci and two other pacakges containing 1.1 Ci. The 12 packages were listed all on one line in the RSR, instead of on 12 separate lines. This apparently confused the burial site personnel, who recorded the amount of activity as though only one 10-Ci package was received.

Mr. Lorenz said that their records did not say exactly what form the waste took, but they are confident it was one of the three following:

1. HTO mixed with concrete.

2. Molecular sieves containing sorbed T_2O/HTO.

3. ZrT or TiT targets.

If it was HTO mixed with concrete, the concrete would have been in 5-gal paint cans. Both the molecular sieves and the targets would have been in pieces of cast iron pipe capped off at both ends. Since the packages were 1 ft^3 in volume, the pipes may have been placed in paint cans. The exact type of closure used for the paint cans is not known, but it would not have been anything special such as soldered or welded.

In the August 10 telephone conversation, Mr. Lorenz described the shipments to Trench 2. The package descriptions are given below:

1. RSR 11212

83 Ci T_2O/HTO on molecular sieves in an iron pipe closed by a pipe nipple at both ends. The pipe was shipped in a fiberboard box.

2. RSR 11211

This shipment contained five packages with >1 Ci H-3:

(a) 110 Ci was the same as described for RSR 11212 above, except that the pipe was placed in a paint can.

(b) 10 Ci was in the form of a metal tritide target. Its primary container is not known, but the secondary container was a fiberboard box.

(c) Three packages, 1.25, 1.26, and 2.0 Ci, listed as containing H-3 and MF?, were essentially all H-3, and were either pump oil sorbed on vermiculite, or aqueous cleanup residues on clay type absorbent. The containers for all three were fiberboard boxes.

3. RSR 3532

Three packages, containing 5.2 and 1.1 Ci H-3, and 9.8 Ci H-3 with a very small amount of C-14, were all oil sorbed on vermiculite, and the containers for all were fiberboard boxes.

4. Shipment of 10/08/70

One package of 7.0 Ci, listed as H-3 and MFP, was again essentially all H-3, and again pump oil sorbed on vermiculite. The vermiculite was in a paint can which was shipped in a fiberboard box.

One package of 10.1 Ci, containing a target, was similar to that in RSR 11211 described above.

5. RSR 3534

One package contained 24 Ci but the form was not known, except that it was one of the three already described, i.e., T_2O/HTO on molecular sieves, targets, or pump oil on vermiculite. The container was a fiberboard box.

One package of 4 Ci was sorbed liquid, again in a fiberboard box.

DRM/nmy
cc: R. E. Barletta
R. E. Davis
C. R. Kempf
J. F. Smalley

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: August 22, 1983

TO: File

FROM: D. R. MacKenzie

SUBJECT: Shipment of 82 Ci of H-3 and Kr-85 by U.S. Radium Corporation to Trench 2

A shipment by U.S. Radium Corporation on June 27, 1969, was listed as containing 82 Ci of H-3 and Kr-85. U.S. Radium Corporation no longer conducts the type of business which uses H-3, but a former division, now an independent company called Safety Light Corporation, carries on the business and has records going back a number of years. However, their records do not go back to 1969. In telephone conversations on August 17 and 22 with Dr. J. G. MacHutchin, Radiation Safety Officer at Safety Light Corporation, it was established that U.S. Radium Corporation never had a plant in Chicago, the point where the shipment originated, so that it must have been sent from the Chicago sales office. Since the normal procedure for a sales office would have been to send such radioactive material back to the plant for disposal, Dr. MacHutchin felt the shipment was made when the Chicago office was being closed down.

It would be rather unlikely that anyone can be found who would know exactly what was in the shipment. However, knowing the types of products being marketed at the time, Dr. MacHutchin's opinion is that it consisted mainly of Kr-85. The Chicago office was dealing at that time with a railroad company which was interested in safety lights containing Kr-85. U.S. Radium worked on development of a light containing $\geq$5 Ci Kr-85, and a number of prototypes were tested by the railroad company. It did not prove feasible to make a light entirely satisfactory to the railroad, and Mr. MacHutchin felt that the shipment in question probably contained the prototype lights they had been testing.

Products containing smaller amounts of tritium were sold to Wright Paterson Air Force Base from the Chicago office, and some of these product samples on hand at the time the office was closed down probably constituted the H-3 in the June 27, 1969, shipment. Exactly what the form of tritium waste might have been cannot be determined, but it seems likely that its amount was of the order of 10 Ci or less. The remaining activity in the shipment (approximately 70 Ci) will be assumed to have been Kr-85, which is not one of the isotopes of concern to BNL for the Sheffield task.

DRM/nmy

cc: R. E. Barletta
R. E. Davis
C. R. Kempf
J. F. Smalley

ABBOTT

R. L. Fredrickson
Corporate Radiation Protection Officer
Employee Health Department

Abbott Laboratories
14th & Sheridan Road
North Chicago, Illinois 60064

August 23, 1983

Donald R. Mac Kenzie
Nuclear Waste Management Division
Brookhaven National Laboratory
Associated Universities, Inc.
Upton, Long Island, New York 11973

Dear Mr. Mac Kenzie:

This letter is in reply to your letter of August 16 to David Schwarz of Abbott Laboratories. My answers to your two questions are as follows:

1. The tritium waste we shipped out in 1967 was tritium gas which we disposed of when we stopped doing catalytic tritiations. The gas was contained in sealed glass bulbs which were packed in containers described in my records as "2R" type. As I recall these were metal pipes, each end of which was closed with a threaded cap. The threads were coated with a heavy glue as a seal, to be welded shut by California Nuclear prior to burial at Sheffield. Each container held 4.5 Curies of hydrogen-3 (from an original five Curies less decay) at time of shipment.

2. The designation MFP did not carry the usual meaning of "mixed fission products". As I recall, the instructions from California Nuclear, where several different radionuclides were in one container, were to use the designation MFP for simplicity. In general, these wastes would be small quantities of carbon-14, hydrogen-3, sulfur-35, etc., from biological or pharmacological research programs. If any of these "MFP" barrels were listed for large amounts of activity, of the order of hundreds of millicuries, they would probably contain short half-life materials such as iodine-131 from the manufacture of radiopharmaceuticals.

I hope that these answers will be helpful to you in your investigations. If I can be of further help, please contact me by writing or by calling me at (312) 937-5276.

Sincerely yours,

R.L. Fredrickson

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: August 25, 1983

TO: File

FROM: D. R. MacKenzie

SUBJECT: Shipments of H-3 >1 Ci by Purdue University to Trench 2

Purdue University made ten shipments ≥5 Ci of H-3 to Trench 2. These are listed in the following Table.

Table

H-3 Shipment ≥5 Ci by Purdue University to Trench 2

RSR	Date	Package	Amount (Ci)
11245	3/13/69	10	5
		23	1
11247	3/27/69	9	<6
11249	4/24/69	16	5
11250	5/15/69	10	5
11252	6/26/69	3	5
11257	10/30/69	15	5
3671	4/07/70	50	5
3673	4/19/70	19	30
3752	8/28/70	12	25
3656	12/01/70	20	10

The form given for all the packages was solid. Except for shipment 11245, which contained two packages with ≥1 Ci H-3, each shipment contained only one package with a high curie content.

Mr. Gordon Born, the University's Radiological Control Officer, was able to locae their records and gave us the following information (telephone calls of August 16 and August 23, 1983). The 30 Ci shipment (RSR 3673) was recorded as consisting of six targets, and several shipments of 5 Ci each contained one target. From his dicussions with other University personnel who were present at the time of the shipments, he is confident that every package containing 5 Ci or a multiple of 5 Ci consisted of targets. That means that all the packages except that containing 1 Ci in shipment 11245, and possibly the <6 Ci package of shipment 11247, consisted of targets. Total curie amount of the targets was at least 95, and possibly 100, out of the ~102 Ci total H-3.

The form of the H-3 was titanium tritide. The person who used the targets repackaged them after use in the same way as they had been when received. This was in glass jars with plastic screw caps contained inside metal paint cans with the lids hammered down. They were disposed of in this form, inside standard 55-gal drums.

DRM/nmy
cc: R. E. Barletta
R. E. Davis
C. R. Kempf
J. F. Smalley

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: August 25, 1983

TO: File

FROM: D. R. MacKenzie

SUBJECT: Am-241 Shipments From Gamma Industries to Trenches 11 and 24

Gamma Industries made a single shipment of 5 Ci Am-241 to Trench 11 and three shipments totalling 12.6 Ci to Trench 24. The shipments are listed in the Table.

Table

Am-241 Shipments by Gamma Industries to Trenches 11 and 24

Trench	RSR	Date	Amount (Ci)	Container
11	1426	3/17/75	5.0	55-gal drum
24	13652	10/21/75	7.0	55-gal drum
24	00465	2/23/76	3.0	55-gal drum
24	23037	5/04/76	2.6	55-gal drum

Gamma Industries is now a division of Nuclear Systems, Inc. of Baton Rouge. The shipments were made from Houston, and according to Mr. Burk McDuff, national sales coordinator for Nuclear Systems, most of the records that had been at Gamma Industries' Houston Office are now at Baton Rouge (telephone conversations of August 9 and 25). Mr. McDuff found sales records, but not radioactive waste disposal records from the period of the shipments. Before searching further for the disposal records, he had occasion to talk with the Radiation Safety Officer at Houston and with the health physics consultant to Nuclear Systems, Inc., Dr. Pullen. Both were familar with the waste and stated that its chemical form was AmO_2, which is in agreement with the notation on RSR 23037 that the chemical form for that shipment was AmO_2. They did not give him any information on the packaging, but this is probably immaterial due to the very low solubility of AmO_2.

DRM/nmy

cc: R. E. Barletta
R. E. Davis
C. R. Kempf
J. F. Smalley

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: September 1, 1983

TO: File

FROM: D. R. MacKenzie DRM

SUBJECT: Large C-14 Shipments to Trenches 2 and 24

There were no C-14 shipments >1 Ci to Trench 1. Those to Trenches 2 and 24 were made by Mallinckrodt, Amersham/Searle and Pathfinder Laboratories, as listed in the Table. Each of these generators was contacted regarding the waste form and packaging in their shipments.

Pathtinder Laboratories had already been contacted regarding their C-14 shipments to Trenches 14A and 23 (26 Ci in three shipments). The information given at that time is contained in a Memo to File dated March 22, 1983, and attached to the March Monthly Letter Report as Appendix B. William Perry of Pathfinder Laboratories, said in a telephone conversation on August 22, 1983, that the C-14 sent to Trench 24 was the same type as that sent later to Trenches 23 and 14A. The waste form and packaging remained the same throughout the period when they shipped to Sheffield.

The situation with Mallinckrodt was much the same, but less definite because their shipment was 8 yrs earlier and they no longer had records going back that far. In a telephone conversation on August 23, 1983, Dr. R. W. Brown of Mallinckrodt Nuclear stated that he felt that the waste form, and probably also the packaging, would have been very similar to that described to BNL in March (telephone conversation, Dr. R. W. Brown to C. R. Kempf, April 4, 1983). The only way to obtain more definite information would be to find the individuals who generated the waste, and the person who packaged it, and Dr. Brown felt this was not feasible.

The Amersham/Searle shipments were discussed with Linda Bagby of Amersham Corporation during telephone conversations of August 22 and September 1. Questioning personnel still at Amersham who were with the Company at the time of these early large C-14 shipments produced no new information. As far as could be ascertained, these shipments were routine except for the larger than usual amounts. They would not have included large numbers of sources, and probably contained unsold material that was being cleared out. Most, if not all, of the packages would thus consist of labelled organic compounds soluble in aqueous and alcohol solutions, as did the later shipments already discussed with Amersham (Memo to File of March 30, 1983, attached to the March Letter Report as Appendix I). Packaging was also considered likely to be similar.

Table

Shipments of C-14 >1 Ci to Trenches 2 and 24

Trench	Shipper	RSR	Date	Package	Amount (Ci)
2	Mallinckrodt	----	9/10/68	5	<1
				6	<1
				7	<1
				8	<1
				9	<2
2	Amersham/Searle	----	2/04/69	1	6
				2	1
2	Amersham/Searle	4537	10/22/69	1	0.5
				2	1.0
				4	0.5
				5	0.1
2	Amersham/Searle	3763	3/06/70	1	0.7
				2	1.0
				5	1.0
				7	0.3
2	Amersham/Searle	3764	7/17/70	1	0.5
				4	0.5
				5	1.0
				6	0.5
2	Amersham/Searle	3765	10/16/70	3	1.5
				4	0.5
				6	0.5
24	Pathfinder Laboratories	03009	11/11/75	1	<7.0
				2	<7.0
24	Pathfinder Laboratories	01557	3/30/76	1	<2.0
				2	<2.0

DRM/nmy
cc: R. E. Barletta
R. E. Davis
C. R. Kempf
J. F. Smalley

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: July 29, 1983

TO: File

FROM: D. R. MacKenzie DRM

SUBJECT: Waste Shipments From Argonne National Laboratory to Sheffield Trenches

Routine Shipments

During the course of operations at the Sheffield LLW burial site, from late 1966 to early 1978, Argonne National Laboratory (ANL) regularly shipped radioactive waste to the site in 120 ft^3 steel bins. A number of shipments containing eight to ten bins each were sent to the trenches covered by BNL's original Sheffield SOW. Each bin was listed as containing 0.45 Ci of MFP (mixed fission products). Since the curie amounts of the shipments were non-trivial, ANL personnel were questioned regarding the isotopic composition and form of the waste (telephone conversations, D. R. MacKenzie to Lyle Cheever, January 14 and 17, 1983). Mr. Cheever, Manager of ANL's Waste Management Operations Department, checked back in their records for us. He could find nothing in the records to indicate what isotopes may have been present in any specific bin, let alone what quantity of any particular isotope was present. The waste was simply collected from all divisions of ANL, placed in the bins, and shipped when enough bins had been filled. It should have been listed as mixed byproduct material rather than mixed fission products. Mr. Cheever described the bins as follows: dimensions 4' x 5' x 6', made of 16 gauge mild steel, flanged at the tops, and closed by bolting lids to the flanges with gaskets.

Special Shipments to Trench 2

A large number of routine shipments containing several hundred bins were sent to Trenches 1 and 2 during the four years they were operating (from the opening of the site until March 1971). During the period February to May 1970, ANL also shipped some considerably higher level waste in 55-gal drums rather than the large steel bins of the routine shipments. These special shipments appear to have been in two series, one from late February to April 21, with drums containing 3-7 Ci MFP in each drum, and one from April 22 to May 13, with drums containing 14-70 Ci each. Because of the nonroutine nature of these shipments it was considered that ANL would be likely to have more information on them than on the regular shipments of general laboratory waste.

The Waste Management Operations Department at ANL was again approached, and its personnel were able to provide most of the information required on the nature of the waste in the special shipments (telephone conversations of D. R. MacKenzie to Lyle Cheever on June 24, July 18 and July 29, 1983). Since ANL's records did not go back to 1970, we sent Mr. Cheever copies of the microfilms of the following RSRs:

4096 and 4099 (3-7 Ci drums)
4101,4102, 4103, 4104, 4109, 4112, 4117, 4121, and 4122 (14-70 Ci drums)
4125 (routine shipment of bins)

The shipments represented by RSRs 4122 and 4125, one a "hot" drum and one a regular shipment of bins, were made on the same day, May 13, 1970.

Mr. Cheever, with the help of personnel in his department who had been present at the time the special shipments were made, identified the waste as coming from hot cell examination of EBR-1 and/or 2 irradiated driver fuel. What appeared to be two series of shipments (3-7 and 14-70 Ci) could have been one from each reactor, but if so, it was not known which series was from which reactor. The important point is that the waste was, indeed, MFP from irradiated fuel. It also contained source material and SNM (since it was, essentially, irradiated fuel which was contaminating tools, equipment, etc.), and activation products such as Co-60 in the stainless steel cladding component. EBR-1 fuel was metal (fissium) and EBR-2 fuel was UO_2

The packaging of the waste was done as follows. Pieces of Schedule 40 black iron pipe were loaded with waste and capped off. Each pipe was packed centrally in a standard DOT 55-gallon drum and the drum sealed in the usual manner. Because of their high radiation fields, the drums were transported in type B3 shipping casks supplied by NECO.

DRM:cv
cc: R. E. Barletta
R. E. Davis
C. R. Kempf
J. Smalley

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: August 19, 1983

TO: File

FROM: D. R. MacKenzie

SUBJECT: Probable Isotopic Composition of Special Shipments From Argonne National Laboratory to Trench 2

Discussions were held with Dr. L. Neimark of Argonne National Laboratory's Materials Science Division regarding the nature of the waste in ANL's special shipments to Trench 2 (telephone conversations of August 10 and August 15, 1983). After talking with the persons who had prepared the waste for shipment (the same sources of information as used by Lyle Cheever - see Memo to File of July 29, 1983), Dr. Neimark concluded that the waste had come from cleanup of one particular hot cell which had been in operation for some 20 yrs and had been accumulating waste over that period.

The cell had been used for metallographic examination of irradiated reactor fuel elements. No chemical treatment was carried out in the cell, so essentially no salts of the uranium and plutonium in the fuel would have been present. The cell was operated in air atmosphere, rather than inert gas. Thus, the small particles of fuel which contained the waste would be almost entirely in the form of oxide, either as fuel which was orginally UO_2, or as metal fuel oxidized in the air atmosphere. No large pieces of fuel would have been present in the waste because of the accountability requirements, so that the waste consisted of small particles of fuel contaminating tools, equipment, discarded reactor internals, cleaning materials, etc.

The waste would have resulted from examination of irradiated fuel, not only from EBR-1 and EBR-2, but also from ETR, MTR, and probably CP-5, so that most of the material was well aged, and the bulk of the by-product activity would have been due to the longer-lived fission and activation products, Cs-137, Sr-90, and Co-60, probably mostly the fission products. Thus, MFP was a reasonable description of the waste, and "well aged" should be added.

Dr. Neimark did not know how the figures for source material and SNM had been arrived at in filling out the RSRs. All RSRs had the same amounts, namely <5 g SNM and <5 lb source. He agreed that there was no basis for

having the same amounts of source material and SNM associated with 3 Ci of MFP as with 70 Ci. All we can conclude is that most of the drums contained very much less than the amounts listed, so that the total amounts listed for these special shipmens were not meant to be realistic, but gave rather, an extremely conservative upper limit.

DRM/nmy
cc: R. E. Barletta
R. E. Davis
C. R. Kempf
J. F. Smalley

BROOKHAVEN NATIONAL LABORATORY

MEMORANDUM

DATE: October 25, 1983

TO: File

FROM: D. R. MacKenzie

SUBJECT: Clarification of Information Regarding ANL Special Shipment Series of 1968 and 1970

It was pointed out in the September Letter Report that the waste in ANL's 1968 series of special shipments was much more dilute than that in the 1970 series (comparable or smaller total radioactivities in 120 ft^3 bins as compared with 7.3 ft^3 drums). We have obtained information on the 1968 series from Mr. W. H. Kline of ANL (telephone conversation of October 7, 1983). Mr. Kline was in charage of preparing the waste in both the 1968 and 1970 series for shipment. He stated that the waste in these two series came from a general cleanup of the whole ANL site, including, but not restricted to, hot cells. That in the 120 ft^3 bins was considered intermediate level, more highly radioactive than the normal ANL shipments, but not requiring shielding. That in the drums had considerably higher radiation fields and required the heavy shielding of the NECO B-3 shipping casks for shipment. All this waste had been stored because of its high radiation field, and some of it had been stored since the early days of ANL's operation. In general, it was well aged, and thus contained mostly isotopes of reasonably long half-life, such as those we are concerned with in this study. A great deal of it was from hot cells and would contain the longer-lived fission products and activation products mentioned previously (Monthly Letter Report of August 1983, Appendix D). It would hot have contained H-3, C-14, and I-129, unless they were coincidentally mixed with other waste, since such isotopes have almost no radiation field and would not have needed to be stored.

Mr. Kline also clarified the description of the shipping procedure for the hot waste (telephone conversation of October 17). This differs considerably in its detail from that obtained earlier (Letter Report of July 1983, Appendix D), although the general picture is similar. The primary waste containers (MAP tubes) were made of aluminum rather than iron. The aluminum tubes were 2, 4, and 6-in. diameter, and of different lengths, with welded bottoms. The tops snapped on with a press fit so they could be closed remotely. These tubes, filled with waste, had been stored in hot cells and elsewhere until the cleanup. They were loaded remotely into the drums by setting them in on end, as many as the drum would hold -- not one centrally located as

we had been told earlier. The drum lids were of the finger-closure type rather than ring-bolt sealed. The drums were shipped in the NECO B-3 cask, 2 per truckload.

DRM/nmy
cc: R. E. Barletta
R. E. Davis
C. R. Kempf

APPENDIX C

ESTIMATE OF TRITIUM RELEASE FROM A CARBON STEEL DRUM OF LABORATORY TRASH BURIED AT SHEFFIELD

C.1. Gasesous Release

In order to estimate the tritium release to trenches from carbon steel drums containing miscellaneous laboratory trash, two mechanisms appear to be important. First, there is release of tritium due to gaseous transport and, secondly, release due to leaching. For drums emplaced at Sheffield, both mechanisms will operate immediately upon emplacement only for those drums breached during emplacement. In this case, it has been conservatively assumed, given the high mobility of tritium, that the entire drum inventory is released during the first year of burial. Based on an assessment using photographs of low-level waste drums in trenches (Section 4.1.2), it is expected that 15% of the drums buried at Sheffield will have been breached.

For the remaining 85%, the drums will initially provide a barrier to liquid. Hence, until the drums are breached due to corrosion, only gaseous release of tritium is expected. This may be estimated from the rate of tritium release from tritiated organic materials. This rate of release is assumed to be the same as that estimated from data for the rate of tritiated gas release from luminous paints.[1] It is also assumed that, on the timescale of interest (i.e., years), the drums present no barrier to gaseous release from this waste even when they have not been breached by corrosion. Accordingly, the rate of gaseous release from this waste (R_g) may be expressed as

$$R_g = k_g\, I(t) \tag{1}$$

where k_g is the rate constant for gaseous tritium release ($k_g = 4.5 \times 10^{-2}\ yr^{-1}$) (Section 4.4.5) and $I(t)$ is the amount of tritium left in the waste form at time t. The quantity $I(t)$ may be determined by solving the following differential equation (which takes radioactive decay into account)

$$-\frac{dI(t)}{dt} = (k_g + \lambda)\, I(t) \tag{2}$$

where λ is the radioactive decay rate constant. Thus, when loss of tritium occurs only through gaseous release, R_g is determined by

$$R_g = I_o\, k_g \exp[-(k_g + \lambda)t] \tag{3}$$

where I_o is the amount of tritium initially in the waste.

C.2. Release Due to Leaching

Once corrosion has breached the drum, leaching can occur, in addition to gaseous release. In reality, these two mechanisms (leaching and gaseous release) are coupled, but since it is not possible to determine the extent of interaction between them, it is assumed that the total rate of loss of tritium from this waste (R_T) is just the sum of the rate of loss due to radioactive decay (R_D) plus that due to gas release (R_g) plus that due to leaching (R_L), i.e.,

$$R_T = R_D + R_g + R_L \tag{4}$$

Since (see Equation 2)

$$R_D + R_g = (k_g + \lambda)\ I(t) \tag{5}$$

Equation 4 may be rewritten as

$$R_T = (k_g + \lambda)\ I(t) + R_L \tag{6}$$

To estimate the loss due to leaching, it is first necessary to make assumptions concerning the lifetime of a carbon steel drum, as well as the induction period of the start to corrosion. A detailed study of the corrosion of steels in soils has been performed by Romanoff.[2] According to this study, the Sheffield site lies in a region identified as Prairie Soils. Since there were no sampling sites in Illinois, sites in neighboring states lying in the same soil group were reviewed. Data from the selected sites indicate a soil pH ranging from 4.6 to 7.0 and soil resistivity in the range of 1000 to 2400 ohm-cm. It is noteworthy that the sampling depth for the Romanoff data was 5 ft or less, whereas, trench depth at Sheffield can be >20 ft. Piciulo et al.[3] report data on various soil samples from an area adjacent to the Sheffield site. The pH of the soils sampled to 33 ft ranged from 6.9 to 7.6, and the resistivity ranged from 4000 to 6500 ohm-cm. These data would indicate large differences between the Sheffield soils and those of the Romanoff study. However, since all soils removed from the trench were probably used as backfill and the presence of the waste itself can affect corrosion of containers, it is assumed that the corrosion data for the selected sites from the Romanoff study may provide a suitable approximation for estimating failure time of a 55-gal sealed drum.

The ranges of corrosion rates for ferrous pipe given by Romanoff for the selected soils are for weight loss, 0.24 to 0.85 oz/ft^2/yr, which converts to 0.37 to 1.30 mil/yr and for pit penetration, 4.4 to 11.8 mil/yr. Assuming uniform corrosion of a 55-gal mild steel drum (50-mil wall thickness), total disintegration of the drum is estimated to occur betwen 38 and 135 yrs. A period from burial to first breach of the drum can be estimated from the pitting data cited above. Penetration of the drum may first occur sometime between 4.7 yrs and 11.4 yrs.

From these results, the period of time before which no breach of the drum occurs has been conservatively estimated to be ~5 years. Further, the mean time to total disintegration of a carbon steel drum has been estimated to be ~86 yrs after the induction period.

For an intact 55-gal drum containing waste, it is assumed that, as soon as an area of the drum disintegrates due to corrosion, all of the tritium contained by that area is immediately leached. Thus, R_L may be expressed as

$$R_L = \frac{I(t)\ \frac{ds(t)}{dt}}{A - s(t)} \tag{7}$$

where ds(t)/dt is the rate of growth of the disintegrated area [s(t)], and A is the surface area of a drum. The course of the drum's corrosion is modelled by assuming that the drum is breached by a single circular pit at t = 5 yr. Furthermore, it is assumed that the radius of this pit (r_p) grows linearly with time, i.e.,

$$r_p = Kt \tag{8}$$

where K is a constant. Thus, since

$$s(t) = \pi r_p^2 = \pi K^2 t^2 \tag{9}$$

the quantity ds(t)/dt becomes

$$ds(t)/dt = 2\pi K^2 t \tag{10}$$

and Equation 7 becomes

$$R_L = \frac{I(t)\ (2\pi K^2 t)}{A - \pi K^2 t^2} \tag{11}$$

Substituting Equation 11 into Equation 6 gives

$$R_T = \frac{dI(t)}{dt} = I(t)\left(k_g + \lambda + \frac{2\pi K^2 t}{A - \pi K^2 t^2}\right) \tag{12}$$

which may be solved to give the amount of tritium left in the waste as a function of time [I(t)]

$$I(t) = I_o \exp\left[-\left((k_g + \lambda)t + \ln\left(\frac{A}{A - \pi K^2 t^2}\right)\right)\right] \tag{13}$$

Since a 5-yr induction period is required for corrosion to breach the drum, i.e., K = 0 for $t \leq 5$ yr, Equation 13 should be rewritten as

$$I(t) = I_o \exp[-(k_g + \lambda)t] \tag{14}$$

for $t \leq 5$ yrs, and

$$I(t) = I(t = 5\ \text{yr})\ \exp\left[-\left((k_g + \lambda)\ (t - 5\ \text{yr}) + \ell n\left(\frac{A}{A - \pi K^2(t - 5\ \text{yr})^2}\right)\right)\right] \quad (15)$$

for $t > 5$ yr. Since

$$I(t = 5\ \text{yr}) = I_o\ \exp[-(k_g + \lambda)(5\ \text{yr})] \quad (16)$$

Equation 15 becomes

$$I(t) = I_o\ \exp\left[-\left((k_g + \lambda)t + \ell n\left(\frac{A}{A - \pi K^2(t - 5\ \text{yr})^2}\right)\right)\right] \quad (17)$$

for $t > 5$ yr. (Note that Equation 14 may be rewritten as

$$I(t) = I_o\ E_\lambda E_g \quad (18)$$

where $E_\lambda [= \exp(-\lambda t)]$ is an exponential term due to radioactive decay and $E_g [= \exp(-k_g t)]$ is an exponential term due to gas release. Also note that Equation 17 may similarly be recast as

$$I(t) = I_o E_\lambda E_g E_L \quad (19)$$

where $E_L = \exp\left(-\ell n\left(\frac{A}{(A - \pi K^2(t - 5\ \text{yr})^2}\right)\right)$ is an exponential term due to leaching.

Since the rate of release of tritium from this waste $R_R(t)$ is defined as

$$R_R(t) = R_g = k_g I(t) \quad (20)$$

for $t \leq 5$ yr and

$$R_R = R_g + R_L = \left(k_g + \left(\frac{2\pi K^2 t}{A - \pi K^2 t}\right)\right) I(t) \quad (21)$$

for $t > 5$ yrs, Equations 14 and 17 may now be substituted into Equations 20 and 21, respectively, to give

$$R_R(t) = k_g I_o\ \exp[-(k_g + \lambda)t] \quad (22)$$

for $t \leq 5$ yr, and

$$R_R(t) = \left[k_g + \left(\frac{2\pi K^2\ (t - 5\ \text{yr})}{A - \pi K^2(t - 5\ \text{yr})^2}\right)\right] I_o \exp\left[-\left((k_g + \lambda)\ t + \ell n\left(\frac{A}{A - \pi K^2(t - 5\ \text{yr})^2}\right)\right)\right] \quad (2$$

for $t > 5$ yr. The amount of tritium released from the waste [AR(t)] is defined by

$$AR(t) = \int_0^t R_R(z)dz \quad (24)$$

and the cumulative fraction of the tritium released from the waste [(CFR(t))] is defined by

$$CFR(t) = AR(t)/I_o \tag{25}$$

Thus, CFR(t) is given by

$$CFR(t) = \frac{k_g}{k_g + \lambda}\left[1 - \exp(-(k_g + \lambda)t\right] \tag{26}$$

for $t \leq 5$ yr, and CFR(t) is given by

$$CFR(t) = \int_o^t \left[k_g + \left(\frac{2\pi K^2 (z - 5\ \text{yr})}{A - \pi K^2 (z - 5\ \text{yr})^2}\right)\right] \exp\left[-\left((k_g + \lambda)\ z + \ell n\left(\frac{A}{A - \pi K^2 (z - 5\ \text{yr})^2}\right)\right)\right] dz \tag{27}$$

for $t > 5$ yr.

3. Estimate of Release as a Function of Time

Equations 26 and 27 may now be used to estimate the cumulative fraction of the tritium released from a drum as a function of time. As mentioned previously, the value of k_g used in this calculation is assumed to be the same as the release rate constant calculated from data given in Reference 1 for tritiated paint (i.e., $k_g = 4.5 \times 10^{-2}\ yr^{-1}$). Additionally, a value for K may be estimated by assuming that the drum has an 86-yr lifetime after it is first breached. Equation 9 may then be used to determine K, i.e.,

$$A = \pi K^2 (86\ \text{yr})^2 \tag{28}$$

and

$$K^2 = \frac{A}{\pi (86\ \text{yr})^2} \tag{29}$$

Substituting the value of k_g given above along with the value of λ for tritium ($5.6 \times 10^{-2} yr^{-1}$) into Equation 26 gives

$$CFR(t) = 0.45\ [1 - \exp\ (-1.01 \times 10^{-1}\ t)] \tag{30}$$

which may be used to calculate CFR as a function of time for $t \leq 5$ yr. Additionally, substituting these same values of k_g and λ along with Equation 29 into Equation 27 gives

$$CFR(t) = \int_0^t \left[4.5 \times 10^{-2} + \left(\frac{2.7 \times 10^{-4}(z - 5\ yr)}{1 - \left(\frac{z - 5\ yr}{86} \right)^2} \right) \right] \exp \left[-\left((1.0 \times 10^{-1} z) - \ln \left(1 - \left(\frac{z - 5\ yr}{86} \right)^2 \right) \right) \right] dz \quad (31)$$

which may be used to calculate CFR as a function of time for t >5 yr.

Table C-1 gives values calculated for the cumulative fractions released due to gas release [$CFR_g(t)$], leaching [$CFR_L(t)$], and to both of these mechanisms combined [$CFR_T(t)$] for each year up to t = 90 yr. For $t \leq 5$ yr, $CFR_g(t)$ and $CFR_T(t)$ were calculated using Equation 30. For t > 5 yr, Equation 31 was used to calculate $CFR_T(t)$; $CFR_g(t)$ was calculated by

$$CFR_g(t) = 4.5 \times 10^{-2} \int_0^t \exp \left[-\left((1.01 \times 10^{-1} z) - \ln \left(1 - \left(\frac{z - 5\ yr}{86} \right)^2 \right) \right) \right] dz \quad (32)$$

and $CFR_L(t)$ was calculated by

$$CFR_L(t) = \int_0^t \left[\frac{2.7 \times 10^{-4}(z - 5\ yr)}{1 - \left(\frac{z - 5\ yr}{86} \right)^2} \right] \exp \left[-\left((1.01 \times 10^{-1} z) - \ln \left(1 - \left(\frac{z - 5\ yr}{86} \right)^2 \right) \right) \right] dz \quad (33)$$

The integrals in Equations 31, 32, and 33 were determined by numerical integration using Simpson's rule with a step size of 0.1 yr. Table C-1 also contains values for the incremental fraction released from the waste [IFR(t)] calculated according to

$$IFR(t) = CFR_T(t) - CFR_T\ (t - 1\ yr) \quad (34)$$

A review of Table C-1 indicates that leaching never becomes an important release mechanism relative to gas release and that, in fact, ~97% of the tritium which is released is lost due to gas release. Table C-1 also demonstrates that, because of radioactive decay, 90% of the tritium which is released is lost during the first 25 yrs (i.e., during the tritium's first two radioactive decay half-lifes).

Table C.1

Cumulative Releases and Total Incremental Fraction Releases of Tritium From Packages Containing Laboratory Trash

t(yr)	$CFR_g(t)$	$CFR_L(t)$	$CFR_T(t)$	IFR(t)
1.0	.42318E-01	0.	.42318E-01	.42318E-01
2.0	.81225E-01	0.	.81625E-01	.38908E-01
3.0	.11637E+00	0.	.11637E+00	.35145E-01
4.0	.14812E+00	0.	.14812E+00	.31747E-01
5.0	.17679E+00	0.	.17679E+00	.28677E-01
6.0	.20270E+00	.70006E-04	.20277E+00	.25979E-01
7.0	.22609E+00	.23434E-03	.22637E+00	.23600E-01
8.0	.24721E+00	.59871E-03	.24780E+00	.21433E-01
9.0	.26627E+00	.99665E-03	.26726E+00	.19459E-01
10.0	.28347E+00	.14591E-02	.28492E+00	.17661E-01
11.0	.29898E+00	.19697E-02	.30095E+00	.16025E-01
12.0	.31297E+00	.25151E-02	.31549E+00	.14537E-01
13.0	.32559E+00	.30835E-02	.32867E+00	.13183E-01
14.0	.33696E+00	.36655E-02	.34062E+00	.11952E-01
15.0	.34720E+00	.42532E-02	.35146E+00	.10833E-01
16.0	.35643E+00	.48400E-02	.36127E+00	.98101E-02
17.0	.36474E+00	.54205E-02	.37016E+00	.88922E-02
18.0	.37223E+00	.59906E-02	.37822E+00	.80582E-02
19.0	.37896E+00	.65467E-02	.38551E+00	.72914E-02
20.0	.38502E+00	.70863E-02	.39211E+00	.66000E-02
21.0	.39047E+00	.76074E-02	.39808E+00	.59725E-02
22.0	.39538E+00	.81084E-02	.40348E+00	.54034E-02
23.0	.39978E+00	.85884E-02	.40837E+00	.48872E-02
24.0	.40374E+00	.90469E-02	.41279E+00	.44192E-02
25.0	.40730E+00	.94833E-02	.41679E+00	.39950E-02
26.0	.41050E+00	.98978E-02	.42040E+00	.36106E-02
27.0	.41337E+00	.10291E-01	.42366E+00	.32623E-02
28.0	.41594E+00	.10662E-01	.42661E+00	.29469E-02
29.0	.41825E+00	.11012E-01	.42927E+00	.26613E-02
30.0	.42033E+00	.11342E-01	.43167E+00	.24028E-02
31.0	.42219E+00	.11652E-01	.43384E+00	.21689E-02
32.0	.42385E+00	.11943E-01	.43580E+00	.19572E-02
33.0	.42535E+00	.12216E-01	.43756E+00	.17657E-02
34.0	.42668E+00	.12471E-01	.43915E+00	.15926E-02
35.0	.42788E+00	.12710E-01	.44059E+00	.14360E-02
36.0	.42895E+00	.12933E-01	.44188E+00	.12945E-02
37.0	.42991E+00	.13141E-01	.44305E+00	.11667E-02
38.0	.43077E+00	.13335E-01	.44410E+00	.10512E-02
39.0	.43153E+00	.13516E-01	.44505E+00	.94668E-03
40.0	.43222E+00	.13684E-01	.44590E+00	.85269E-03
41.0	.43283E+00	.13840E-01	.44667E+00	.76806E-03
42.0	.43338E+00	.13985E-01	.44736E+00	.69092E-03
43.0	.43386E+00	.14120E-01	.44798E+00	.62168E-03
44.0	.43430E+00	.14245E-01	.44854E+00	.55923E-03
45.0	.43468E+00	.14360E-01	.44904E+00	.50291E-03

Table C.1, Continued

Cumulative Releases and Total Incremental Fraction Releases of Tritium From Packages Containing Laboratory Trash

t(yr)	$CFR_g(t)$	$CFR_L(t)$	$CFR_T(t)$	IFR(t)
46.0	.43503E+00	.14467E-01	.44950E+00	.45213E-03
47.0	.43534E+00	.14567E-01	.44990E+00	.40636E-03
48.0	.43561E+00	.14658E-01	.45027E+00	.36512E-03
49.0	.43585E+00	.14743E-01	.45060E+00	.32796E-03
50.0	.43607E+00	.14821E-01	.45089E+00	.29450E-03
51.0	.43626E+00	.14894E-01	.45115E+00	.26437E-03
52.0	.43643E+00	.14961E-01	.45139E+00	.23725E-03
53.0	.43658E+00	.15022E-01	.45160E+00	.21284E-03
54.0	.43672E+00	.15079E-01	.45180E+00	.19088E-03
55.0	.43684E+00	.15132E-01	.45197E+00	.17113E-03
56.0	.43694E+00	.15180E-01	.45212E+00	.15337E-03
57.0	.43703E+00	.15225E-01	.45226E+00	.13741E-03
58.0	.43712E+00	.15265E-01	.45238E+00	.12306E-03
59.0	.43719E+00	.15303E-01	.45249E+00	.11018E-03
60.0	.43725E+00	.15338E-01	.45259E+00	.98601E-04
61.0	.43731E+00	.15370E-01	.45268E+00	.88208E-04
62.0	.43736E+00	.15399E-01	.45276E+00	.78880E-04
63.0	.43740E+00	.15426E-01	.45283E+00	.70510E-04
64.0	.43744E+00	.15451E-01	.45289E+00	.63001E-04
65.0	.43747E+00	.15474E-01	.45295E+00	.56208E-04
66.0	.43750E+00	.15495E-01	.45300E+00	.50233E-04
67.0	.43753E+00	.15514E-01	.45304E+00	.44824E-04
68.0	.43755E+00	.15532E-01	.45308E+00	.39979E-04
69.0	.43757E+00	.15548E-01	.45312E+00	.35640E-04
70.0	.43759E+00	.15563E-01	.45315E+00	.31756E-04
71.0	.43760E+00	.15576E-01	.45318E+00	.28281E-04
72.0	.43761E+00	.15589E-01	.45320E+00	.25172E-04
73.0	.43762E+00	.15600E-01	.45322E+00	.22392E-04
74.0	.43763E+00	.15611E-01	.45324E+00	.19908E-04
75.0	.43764E+00	.15620E-01	.45326E+00	.17688E-04
76.0	.43765E+00	.15629E-01	.45328E+00	.15706E-04
77.0	.43765E+00	.15637E-01	.45329E+00	.13937E-04
78.0	.43766E+00	.15645E-01	.45330E+00	.12358E-04
79.0	.43766E+00	.15652E-01	.45332E+00	.10950E-04
80.0	.43767E+00	.15658E-01	.45332E+00	.96952E-05
81.0	.43767E+00	.15663E-01	.45333E+00	.85773E-05
82.0	.43767E+00	.15669E-01	.45334E+00	.75817E-05
83.0	.43767E+00	.15673E-01	.45335E+00	.66958E-05
84.0	.43768E+00	.15678E-01	.45335E+00	.59077E-05
85.0	.43768E+00	.15682E-01	.45336E+00	.52072E-05
86.0	.43768E+00	.15685E-01	.45336E+00	.45849E-05
87.0	.43768E+00	.15689E-01	.45337E+00	.40324E-05
88.0	.43768E+00	.15692E-01	.45337E+00	.35422E-05
89.0	.43768E+00	.15695E-01	.45337E+00	.31076E-05
90.0	.43768E+00	.15697E-01	.45336E+00	.27225E-05

C.4 References

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2. M. Romanoff, Underground Corrosion, NBS Circular 579 PB-168350, April 1957.

3. P. L. Piciulo, C. E. Shea, and R. E. Barletta, "Analysis of Soils From an Area Adjacent to the Low-Level Radioactive Waste Disposal Site at Sheffield, Illinois," Draft Report, BNL-NUREG-31667, 1982.

APPENDIX D

ISOTOPIC RELEASE INFORMATION

The following tables are on the microfiche card attached to the inner rear cover of the report.

Table D.1 - Tritium IFRs and CFRs by Case

Table D.2 - Tritium Incremental and Cumulative Releases by Trench

Table D.3 - Fuel Cycle IFRs and CFRs for Cs, Sr, and Co by Case

Table D.4 - Fuel Cycle Incremental and Cumulative Releases for Cs, Sr, and Co by Trench

Table D.5 - Non-Fuel Cycle Incremental and Cumulative Releases for Cs and Sr by Trench

Table D.6 - Total Incremental and Cumulative Releases for Cs, Sr, and Co by Trench

APPENDIX E

CALCULATION OF PARAMETERS SPECIFIC TO THE "RINSE" MODEL, AND DERIVATION OF THE EXPRESSION FOR IFR

E.1 Rinse Volumes

For each drum angular orientation, the amount of rainfall which succeeds in entering the inner lead lining is calculated from:

$$A \cdot \cos \alpha \cdot r(I,II,III)$$

where

- A is the area of the opening around the lead lining lid (30.4 cm^2)
- $\cos \alpha$ is the cosine of the angle (α) at which the drum is "tilted" in the trench (upright is taken as $\alpha = 0^o$, horizontal is taken as $\alpha = 90^o$)
- $r(I,II,III)$ is the respective annual rainfall column height for rainfall I (89 cm/yr), rainfall II (48.3 cm/yr), and rainfall III (6.4 cm/yr).

E.2 Standing Bath Volumes

The 3M lead-lined drums have been assumed to have 50% void space in the inner lead lining and thus to have 50% of the total cylindrical void available for containment of the rainwater which manages to enter the lining. This rainwater accumulates to create what has been termed the "standing bath" in the liner. In calculation of the bath volume, use has been made of the dimensions given on the engineering diagram, Figure 4.1. The size of the bath varies with the angle of tilt in the trench, as mentioned earlier. A sample calculation for the bath volume for a 30^o to upright drum is given below. Based on geometry, the total bath volume that would be possible to fit into one of these drums tilted at such an angle consists of two solid forms: (1) a cylinder, and (2) half cylinder (a cylinder with a diagonal plane bisecting it). The drums tilted from upright will be able to contain water up to a level at which the water can exit the drum. The level sought by the water produces a plane horizontal to the ground surface and it is this plane which, in a tilted drum, bisects the second cylinder, (2), above. The first cylinder, (1), represents the filled "base" of the tilted drum. The heights (and, hence, volumes) of these two forms vary with the drum tilt angle. The height of cylinder (1) is greater than that of half-cylinder (2) for small angles of tilt (with respect to the upright position) while as the angle increases (approaching 90^o or horizontal in the trench), the height of half-cylinder (2) gets successively larger than that of cylinder (1).

CALCULATION OF PARAMETERS SPECIFIC TO THE "RINSE" MODEL, AND DERIVATION OF THE EXPRESSION FOR IFR

For a drum tilted at 30° to upright, the bath volume would be:

$$V_{30^\circ} = F[r_1^2 \pi h_1 + \frac{1}{2} r_1^2 \pi h_2] \qquad (E.1)$$

where

V_{30° is the standing bath volume for a 30° tilt drum,

F is the free space fraction (0.5 assumed)

r_1 is the radius of the lining internal cylinder volume (6.3 in.)

π is the constant pi

h_1 is the height of the "base" cylinder (16.0 in.), and

h_2 is the height of the horizontal-plane-biscected cylinder (6.3 in.)

Conversion of inches to centimeters and substitution of the appropriate values yields a volume of 1.9×10^4 cm^3 for the standing bath in a drum tilted at 30° to upright, assumed to have 50% free space.

E.3 Derivation of Incremental Fraction Release Formula for the "Rinse" Model

The amount of material acquired by the rinse is simply the product of the rinse volume and the waste "concentration" of the standing bath. This is represented by:

$$A_1 = \frac{\ell x_o}{Q} \qquad (E.2)$$

where

A_1 is the amount of material acquired by the first rinse (equivalently the amount of material lost by the "bath"),

ℓ is the rinse volume (cm^3)

x_o is the initial "source" or waste material amount (Ci),

and Q is the standing bath volume.

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It can be seen that x_o/Q is, essentially, the "initial activity concentration" in the standing bath at the end of the "induction" (or rainwater accumulation) period and, just prior to the first rinse. To be consistently conservative, it has been assumed that the rinse volume does not, during the rinse, contribute to the standing bath volume.

By the time of the second rinse (i.e., the second year following the induction period), the standing bath "source" will have been decreased by the amount of material lost in the first rinse, that is,

$$S_2 = x_0 - \frac{\ell x_0}{Q} \qquad (E.3)$$

where

S_2 is the "source" in the waste for the second year (Ci), and all other variables have been previously defined. The amount of material lost to the second rinse is then the product of the rinse volume and this new "source" concentration:

$$\frac{\ell \left(x_0 - \frac{\ell x_0}{Q} \right)}{Q}$$

where

ℓ is the rinse volume,

and

$\left(x_0 - \frac{\ell x_0}{Q} \right)/Q$ is the second rinse "source" concentration.

This process of successively decreasing the source concentration and then finding the amount acquired by the rinses leads to, for example, the expression below for the amount of material lost in the fifth rinse:

$$A_5 = x_0 \left[\frac{\ell}{Q} - 4\left(\frac{\ell}{Q}\right)^2 + 6\left(\frac{\ell}{Q}\right)^3 - 4\left(\frac{\ell}{Q}\right)^4 + \left(\frac{\ell}{Q}\right)^5 \right]. \qquad (E.4)$$

The ratio of rinse volume to standing bath volume, ℓ/Q, has been called z. Through substitution of z for ℓ/Q and then factoring out one z, the bracketed portion of the equation can be seen to have coefficients that correspond to

CALCULATION OF PARAMETERS SPECIFIC TO THE "RINSE" MODEL, AND DERIVATION OF THE EXPRESSION FOR IFR

the sixth line of the binominal expansion of the expression (1 -z). Thus, the generalized bracketed portion becomes $(1 - z)^{n-1}$ where n is the number of the rinse and, for the fifth rinse, the amount of material released is:

$$A_5 = x_0\, z(1 - z)^{5-1} = x_0\, z(1 - z)^4. \qquad (E.5)$$

The incremental fraction released can thus be written as:

$$IFR = z(1 - z)^{n-1}, \qquad (E.6)$$

and, for calculation of actual activity amounts, the "source", x_0, must be included. It is understood that x_0 must be taken individually as Cs-137 and Sr-90 for the 3M lead-lined drum wastes and, also, must be decay-corrected according to

$$x_0 = x_0^1 \exp(-\lambda t) \qquad (E.7)$$

where

x_0 is the decay-corrected waste activity (Cs-137 and Sr-90) "source" (Ci)

x_0^1 is the initial waste activity (Cs-137 and Sr-90) "source" (Ci)

λ is the decay constant for the individual radioisotope considered (0.023 yr^{-1} for Cs-137 and 0.025 yr^{-1} for Sr-90)

and

t is the time of decay (yr).

The wastes have been assumed to be totally soluble and homogeneously mixed in the standing bath.

E.4 Induction Period Calculation

For a given rainfall, the "induction" period, during which the rainwater accumulates in the lead liner, can be calculated as:

$$T = V_{SB}/V_{r_{(I,II,III)}}$$

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where

T is the time to "bath" filling (yrs),

V_{SB} is the standing bath volume (cm^3)

$V_{r(I,II,III)}$ is the volume of the annual rinse for rainfall I,II, or III (cm^3).

These 3M lead-lined packages have been assumed to be partitioned as 1/3:1/3:1/3 and subjected to rainfalls I, II, and III. Thus, there are three separate induction periods and, consequently three separate initial times of release (the first release can only occur, according to the model, after the standing bath has reached the level of exit from the drums).

Actual calculated values of the two release model parameters are summarized in Table E.1.

Table E.1

Values for Release Model Parameters

	Drum Orientation					
	Upright	30°	45°	60°	(on side) 90°	Upside Down
"Area" Exposed to Rainfall) (cm^2)	30.4	26.3	21.5	15.2	15.2	7.92 E2 or 4.43 E2
Volume "Bath"	2.24 E 4	1.93 E 4	1.68 E 4	1.16 E 4	1.12 E 4	2.24 E4
Rinse Volume I (cm^3)	2.71 E 3	2.43 E 3	1.91 E 3	1.35 E 3	1.35 E 3	7.05 E4 or 3.95 E4
Z_I	1.21 E-1	1.21 E-1	1.14 E-1	1.17 E-1	1.21 E-1	
Rinse Volume II (cm^3)	1.47 E 3	1.27 E 3	1.04 E 3	7.34 E 2	7.34 E 2	3.83 E4 or 2.14 E4
Z_{II}	6.58 E-2	6.58 E-2	6.18 E-2	6.33 E-2	6.55 E-2	
Rinse Volume III (cm^3)	1.83 E 2	1.67 E 2	1.37 E 2	9.65 E 1	9.65 E 1	5.03 E3 or 2.81 E3
Z_{III}	8.62 E-3	8.65 E-3	8.13 E-3	8.32 E-3	8.62 E-3	
Induction Period to "Bath" Fill	"Bath" Volume (50%) / Rinse Volume					
T_I (year)	8.27	8.25	8.80	8.59	8.36	
T_{II} (year)	1.52 E 1	1.52 E 1	1.62 E 1	1.58 E 1	1.53 E 1	
T_{III} (year)	1.16 E 2	1.16 E 2	1.23 E 2	1.20 E 2	1.16 E 2	

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13 ABSTRACT (200 words or less)

An inventory has been compiled of the isotopes of half-life >5 years buried in eight of the trenches at the Sheffield low-level waste burial site. This was accomplished through a search of the Sheffield radioactive shipment records (RSRs). Pertinent information from some 3200 fuel cycle RSRs and 1700 non-fuel cycle RSRs has been stored in a computerized data base and used to develop the inventory. Results of the compilation are in disagreement with the two previous estimates for H-3 and for C-14. In particular, non-fuel cycle H-3 inventory for the eight trenches of the present study is approximately a factor of 2 higher than either previous estimate of total site inventory. Modeling of release processes has been carried out in order to obtain estimates of isotopic release rates from waste packages to the trenches. This modeling is highly speculative, but believed to be state-of-the-art. It required information not only on amounts of the different isotopes, but also on the waste forms and containers holding them. Such information was generally not given on the RSRs and had to be obtained by contact with the generators. Estimated numerical release rate data are given for each trench for H-3, C-14, Cs-137, Sr-90, and Co-60. I-129 is expected to have been totally released within a year of container breaching by corrosion. Most of the Pu, in the form of oxide, will probably not be released at a significant rate.

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