



ORAU TEAM Dose Reconstruction Project for NIOSH

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ACRONYMS AND ABBREVIATIONS

BEST	Bioassay Enrollment Scheduling and Tracking (System)
cpm	counts per minute
d	day
DOE	U.S. Department of Energy
dpm	disintegrations per minute
EEOICPA	Energy Employees Occupational Illness Compensation Program Act of 2000
g	gram
HPC	Health Physics Checklist
hr	hour
IVML	In-Vivo Measurements Laboratory System
LANL	Los Alamos National Laboratory
L	liter
MDA	minimum detectable activity
MDC	minimum detectable concentration
min	minute
ml	milliliter
nCi	nanocurie
NDA	nondetectable activity
NIOSH	National Institute for Occupational Safety and Health
ORAU	Oak Ridge Associated Universities
pCi	picocurie
RaLa	radioactive lanthanum
STEXT	Standard Text
TIB	technical information bulletin
TUPo	tritium, uranium, polonium
UNAA	Uranium Neutron Activation Analysis
U.S.C.	United States Code
VAX	Virtual Address Extension
μCi	microcurie
μg	microgram
§	section or sections

1.0 INTRODUCTION

Technical information bulletins (TIBs) are not official determinations made by the National Institute for Occupational Safety and Health (NIOSH) but are rather general working documents that provide historic background information and guidance to assist in the preparation of dose reconstructions at particular sites or categories of sites. They will be revised in the event additional relevant information is obtained about the affected site(s). TIBs may be used to assist NIOSH staff in the completion of individual dose reconstructions.

In this document, the word “facility” is used as a general term for an area, building, or group of buildings that served a specific purpose at a site. It does not necessarily connote an “atomic weapons employer facility” or a “Department of Energy (DOE) facility” as defined in the Energy Employees Occupational Illness Compensation Program Act of 2000 [42 U.S.C. § 7384l(5) and (12)].

2.0 PURPOSE

The purpose of this TIB is to describe the information contained in the repository database created for Los Alamos National Laboratory (LANL) bioassay data. The repository database was created through a collaborative effort between the Oak Ridge Associated Universities (ORAU) Team and LANL staff to function as a repository for current and historical bioassay data for LANL workers. It is hosted and maintained by LANL. The ORAU Team has a subset (snapshot) of this database on its network for use in dose reconstruction, coworker studies, or other needs. The focus of this document is the contents of the database used by the ORAU Team and the sources of this information.

3.0 BACKGROUND

Much of the historical bioassay data for LANL workers were archived from legacy computer systems that are no longer used. These obsolete formats made retrieval of this information impracticable, which was an impediment for the LANL staff in its efforts to respond to requests for internal monitoring data to support dose reconstruction under EEOICPA. In particular, bioassay data from before 1991 were either essentially unavailable due to storage on obsolete archival systems, or were stored across numerous ad hoc systems that made them impracticable to retrieve. Given these difficulties, a joint effort was carried out between the ORAU Team and LANL staff to create and populate a single database for historical bioassay data for LANL workers.

Based on a reconnaissance of the various databases and datasets that existed at LANL, it was decided to create the bioassay database by modifying LANL’s existing Bioassay Enrollment, Scheduling, and Tracking (BEST) system. This system already contained bioassay data from approximately 1991 to the present. Modification and adaptation of the BEST system was an iterative process that was carried out in parallel with the retrieval and evaluation of historical bioassay data. This TIB does not describe the modification of the BEST system. Its focus is the historical worker bioassay data incorporated into the database and related information pertinent to internal monitoring of LANL workers.

Beyond that already in the BEST system, the additional data incorporated into the database were derived from three principal sources:

- *In vivo* data from existing and historical databases,
- *In vitro* data for americium and plutonium from a standalone database, and
- *In vitro* data for tritium, uranium, and polonium (TUPo) from an obsolete electronic archive format.

Laboratory notebooks that contained historical bioassay data were also identified and retrieved. These notebooks were used to validate a number of the historical TUPo records, and also to search for data for other analytes of interest such as ^{90}Sr and mixed fission products.

4.0 IN VIVO DATA

4.1 **IN VIVO DATA INCORPORATED INTO THE BIOASSAY DATABASE**

Initially, *in vivo* counting data were in two databases maintained by the LANL In-Vivo Measurements Laboratory (IVML). One of them held data from the 1960s through 2003, and the other held data from 2003 to the present. These two IVML databases were incorporated into the LANL bioassay database, from which the ORAU Team's database was derived.

The older IVML database did not report minimum detectable activity (MDA) values and contained count data only for positive results, i.e., if a count had not indicated positive activity then the record contained only the associated administrative information. The newer IVML database included MDA information from more recent counts. Both of the IVML databases contained records that showed "NDA" (nondetectable activity) or similar annotations, with no corresponding MDA value. It was common practice at LANL to count an individual and then manually look at the spectrum to see if any peaks were present. If peaks were noted, the nuclide was identified and an activity was established. If no peaks were seen it was simply annotated that way (i.e., as NDA).

By far, the majority of the *in vivo* counting records in the ORAU Team's database are counts for ^{241}Am or ^{239}Pu . Of the 106,950 *in vivo* records, 44,653 (41%) are counts for ^{241}Am and 43,648 (41%) are for ^{239}Pu . The next largest contributors to the total number of *in vivo* records are ^{235}U and ^{234}Th . These analytes represent 5,685 and 5,680 records, respectively (5% each). The *in vivo* records in the LANL bioassay database specify 81 different analytes. These include specific radionuclides and other descriptors such as "mixed fission products" or "unidentified." The analytes are shown in Table 4-1 along with the corresponding total number of records, the number of non-null results, if any; and the count date range. The *in vivo* records report a result or a MDA, but not both, i.e., the two are mutually exclusive. Thus, the non-null results are those where something other than a non-detection was reported. There were 28,514 non-null results, i.e., records that reported a value rather than a MDA, among the 106,950 *in vivo* records (27%). Of these, 8,859 were less than zero and 1,123 were zero. It is not clear what was meant by reporting a non-null result for the "Unidentified" analyte or how the count data were converted to activity. The same can be said for the non-null results for "Fission Prods" and "Tungsten." All non-null *in vivo* results and MDAs in the database are reported in nCi.

Table 4-1. Analytes listed for *in vivo* counting records in the ORAU Team's database.

Analyte	Total records	Non-null records	Count date range	
			From	To
AM-241	44653	7651	1/1/1969	2/11/2009
AS-72	3	3	9/14/1988	10/6/1989
BA-140	48	48	6/25/2008	2/2/2009
BE-7	934	186	5/12/1970	2/10/2009
BI-213	14	14	12/22/2004	2/10/2005
BI-214	6	6	1/5/1993	5/10/1993
BR-76	12	5	9/2/1980	11/16/1998
BR-77	17	10	9/2/1980	11/16/1998
C11N13	1308	143	5/12/1970	12/23/2003
CD-109	19	12	10/20/1976	11/16/1998
CE-141	8	1	9/2/1980	11/16/1998
CF-249	9	9	8/1/1979	2/24/1986
CM-244	1	0	3/6/2003	3/6/2003

Analyte	Total records	Non-null records	Count date range	
			From	To
CO-56	1	1	9/16/1982	9/16/1982
CO-57	8	8	3/26/1982	12/7/1989
CO57CO58	1	1	5/23/1979	5/23/1979
CO-58	121	121	9/16/1982	2/10/2009
CO-60	76	63	8/6/1970	9/18/2007
CR-51	1	1	5/16/1985	5/16/1985
CS-134	128	115	8/6/1970	12/4/2006
CS-137	495	482	7/9/1969	2/2/2009
CU-64	1	1	2/9/1990	2/9/1990
CU-67	1	1	1/14/1983	1/14/1983
EU-152	451	27	9/7/1979	12/23/2003
FE-59	1	1	5/24/1985	5/24/1985
Fission prods	1	1	12/13/1972	12/13/1972
GD-146	115	115	12/4/2006	2/10/2009
GD-153	114	114	6/25/2008	2/10/2009
GE67GA67	1	1	12/2/1987	12/2/1987
GE68GA68	5	5	7/1/1985	11/25/1987
HF-173	3	1	3/13/1997	5/14/1999
HF-175	120	120	11/22/2005	2/10/2009
HG-195M	8	1	9/2/1980	11/16/1998
HG-197	154	147	9/2/1980	2/10/2009
HG-197M	8	1	9/2/1980	11/16/1998
HG-203	25	18	9/2/1980	11/16/1998
I-123	2	2	10/4/1984	10/4/1984
I-125	159	159	4/20/1976	5/22/1995
I-131	13	13	1/15/1970	4/3/1985
I-132	1	1	12/21/1989	12/21/1989
LA-140	48	48	6/25/2008	2/2/2009
LU-172	115	115	12/4/2006	2/10/2009
LU-173	114	114	6/25/2008	2/10/2009
Mixed activation products	7	0	3/5/1963	9/20/1963
Mixed fission products	7	0	3/5/1963	9/20/1963
MN-54	774	26	5/12/1970	12/4/2006
NA-22	1178	17	9/20/1979	12/23/2003
NA-24	6	2	5/12/1970	8/15/1975
NB-95	48	48	6/25/2008	2/2/2009
ND-147	8	1	9/2/1980	11/16/1998
OS-185	11	4	9/2/1980	11/16/1998
P-32	2	2	6/13/1972	6/13/1972
PB-212	15	15	3/6/1984	2/10/2005
PB-214	14	14	12/22/2004	2/10/2005
PU-238	110	110	7/17/1969	2/7/2007
PU238PU239	2	2	1/25/1982	2/1/1983
PU-239	43648	6646	1/1/1969	2/11/2009
RA-226	4	4	9/14/1970	10/25/2005
RB-83	13	6	9/2/1980	11/16/1998
RB-84	11	4	9/2/1980	11/16/1998
SB-124	11	4	9/2/1980	11/16/1998
SC-46	8	8	9/16/1982	7/14/1987
SE72AS72	1	1	10/12/1988	10/12/1988
SE-75	32	25	9/2/1980	11/16/1998
SM-145	8	1	9/2/1980	11/16/1998
TA-179	9	2	9/2/1980	11/16/1998
TA-182	114	114	6/25/2008	2/10/2009

Analyte	Total records	Non-null records	Count date range	
			From	To
TE-132	1	1	12/21/1989	12/21/1989
TH-234	5680	5680	1/12/2004	2/11/2009
TL-201	11	4	9/2/1980	11/16/1998
TL-202	14	7	9/2/1980	11/16/1998
Tungsten	1	1	10/8/1993	10/8/1993
U-235	5685	5685	4/9/1975	2/11/2009
U-237	7	7	7/2/1975	2/1/1979
U-238	3	3	10/22/1979	6/21/2000
Unidentified	2	2	1/7/1975	4/6/1976
V-48	4	4	9/16/1982	9/13/1984
YB-169	115	115	12/4/2006	2/10/2009
ZN-65	11	11	7/30/1970	2/9/1990
ZR-95	49	49	10/26/1972	2/2/2009
ZR95NB95	3	3	12/5/1985	1/13/1986

4.2 OTHER SOURCES OF *IN VIVO* DATA

There are *in vivo* measurement data available at LANL in addition to those from the two IVML databases. Data exist from the pre-1960 period, but these counts were not considered to have been routine measurements and therefore were not included with the information in the IVML databases. In addition, some paper records exist, such as *beige cards* and *white cards*, that contain analyst comments on how radionuclide activities were calculated for individuals and physiological information such as chest wall thickness. This supplemental information was not included in the LANL bioassay database.

5.0 IN VITRO DATA

The *in vitro* monitoring data consolidated into the LANL bioassay database came from four principal sources: (1) the existing BEST system, (2) a database of plutonium and americium results back to 1944, (3) an electronic archive of TUPO data, and (4) LANL laboratory notebooks.

The BEST system already contained *in vitro* (urinalysis and fecal) monitoring data for plutonium, americium, tritium, and uranium. These data, from both LANL and vendor laboratories, were believed to be complete back to approximately 1991. These more recent data served as a template for the other sources of *in vitro* monitoring data during the modification of the BEST system to create the LANL bioassay database.

The *in vitro* data in the ORAU Team's LANL bioassay database is stored in two tables: one for "pre-1991" data and one for "post-1991." The 1991 demarcation date is not absolute, as there are some post-1991 data in the pre-1991 table and vice versa. The data fields differ between the two tables, reflecting differences in how LANL recorded and managed bioassay data as its program evolved.

5.1 PRE-1991 *IN VITRO* DATA FOR PLUTONIUM AND AMERICIUM

A large number of historical *in vitro* bioassay records for plutonium and americium had already been entered into a modern database. These records, which covered the period from 1943 through 1990, had been compiled for use in statistical studies and included urine, fecal, tissue (from autopsy), and associated incident-related data. The data were used along with modern data for the statistical assessments.

The historical plutonium and americium records had some overlap with the existing BEST data, so they were checked for duplication before being incorporated into the LANL bioassay database.

Information pertaining to incidents associated with specific bioassay records was included so the LANL database could serve as a consolidated source of historical plutonium and americium data. The ORAU Team's version of the LANL bioassay database contains analysis results for ^{241}Am , ^{238}Pu , and ^{239}Pu in its pre-1991 data table.

The pre-1991 data table contains 58 records for ^{241}Am . The collection dates for these range from 2/17/1975 to 9/1/2000. All were analyzed by radiometric alpha spectrometry, denoted by the analysis type code RAS. 48 of the 58 results are tissue analysis data from the LANL human tissue program. Of the 10 non-tissue analyses, 7 are spot fecal and 3 are simulated 24-hour urine. The collection dates for these range from 6/23/1998 to 9/1/2000. The analysis results are reported in pCi/g. All of the urinalysis results are 0 pCi/g. For the fecal analyses, two of the results are negative and one is zero. No MDCs are given.

The pre-1991 table contains 47,349 records for ^{238}Pu covering a date range from 12/14/1967 to 4/9/2000. 47,287 of the records (99.9%) are urinalyses, 55 are tissue analyses, and 7 are fecal analyses. The collection dates for the 7 fecal analyses match those for the ^{241}Am records.

The collection dates for the ^{238}Pu urinalyses range from 12/14/1967 to 7/4/1992. All results are in pCi/24 hours. 18,006 (38%) of the urinalysis results are negative and 8,582 (18%) are zero. No MDCs are given. For the analysis type the user is referred to (Clark 2005).

The pre-1991 data table contains 91,445 records for ^{239}Pu covering a date range from 8/1/1944 to 4/9/2000. 91,375 (99.9%) of the records are urinalyses, 63 are tissue analyses, and 7 are spot fecal. The collection dates for the 7 spot fecal samples match those for the ^{241}Am and ^{238}Pu analyses.

The collection dates for the ^{239}Pu urinalyses range from 8/1/1944 to 5/13/1998. All results are in pCi/24 hours. 25,315 (28%) of the results are negative and 20,249 (22%) are zero. No MDCs are given. For all but 15 of the ^{239}Pu urinalyses the user is referred to (Clark 2005) for the analysis type. The other 15 show analysis by RAS.

5.2 PRE-1991 *IN VITRO* DATA FOR TUPO

Historical bioassay data for TUPO had been archived from a Digital Equipment Corporation Virtual Address Extension (VAX) computer system to a series of Standard Text (STEXT) files as described by Lawrence (1991). There were four groups of files that contained data for tritium, polonium, ^{235}U , and ^{238}U . The STEXT files had been transferred to LANL's modern file server, but could not be used to generate data to support dose reconstruction because of the obsolete formats. Before migrating the information in these files to the bioassay database, legacy conversion software had to be used to extract the data and write them to spreadsheets. The spreadsheets were then imported into a Microsoft® Access™ database as an interim step in preparing the data for upload to the new database. This interim Access™ database used to manage and consolidate the TUPO data extracted from the VAX files. It should not be confused with the Access™ database used by the ORAU Team.

Documentation for the VAX file data fields was not found. Extracting the data into spreadsheets allowed delineation and definition of the data fields through discussions with LANL staff and examination of laboratory notebooks. The data fields varied from nuclide to nuclide and from file to file. It was necessary to work with each file individually to determine what data were provided and their format. The data field definitions that were determined for each file are presented in subsequent sections that address individual nuclides.

Once the data fields were defined, an interim Access™ database was created to contain the 147,690 records from the archived VAX files. Additional fields were defined to capture any revisions made to the data while preserving the original. These included a "Data_Comments" field to incorporate

comments to explain changes, questions, or other issues associated with the original data. Table 5-1 shows the data fields for the interim Access™ database.

Table 5-1. Definition of fields in Access™ database.

Field name	Data type	Description	Comment
Name	Text	Employee name	
Z_Num	Text	Employee number	Not all VAX data in this field were true Z (employee) numbers. In some cases V (visitor) numbers were still present. In others an alphanumeric identification number was used.
GrpEmp	Text	Employer and group identification	
S_Date	Double	Original bioassay sample date	
Rev_S_Date	Double	Revised bioassay sample date	Revised or corrected date.
S_Type	Text	Bioassay sample type code	
S_Res	Double	Original bioassay sample result	
S_Units	Text	Original bioassay reporting units	
Rev_S_Res	Double	Revised bioassay sample results	Revised or corrected results.
Rev_S_Units	Text	Revised bioassay reporting units	Revised or corrected reporting units.
Report_Res	Double	Reported bioassay results	In some cases bioassay results were converted to and reported in different units than the original data.
Report_Units	Text	Reported bioassay units	
Uncert	Single	Estimate of measurement uncertainty	
Uncert_Units	Text	Uncertainty units	
Uncert_Code	Text	Uncertainty code	Specifies the level of confidence in the uncertainty estimate.
S_MDA	Double	Bioassay sample MDC	
Rev_MDA	Double	Revised bioassay sample MDC	Revised or corrected MDC.
MDA_Units	Text	MDC reporting units	
S_Size	Text	Bioassay sample size	Sample size was rarely reported. Bioassay results were typically normalized to sample volume.
Size_Units	Text	Bioassay sample size units	
Book	Text	LANL Notebook number	Provided notebook and page reference for bioassay results recorded in VAX data files.
Page	Double	Notebook page reference	
Rev_Book	Text	Revised notebook number	Revised or corrected notebook reference.
Rev_Page	Double	Revised page reference	Revised or corrected page reference.
Analyst	Text	Laboratory technician ID (usually initials)	
A_Type	Text	Code specifying analytical method	
Book_Comments	Text	Original comments from laboratory notebook	Limited data field space restricted frequent incorporation of laboratory notebook comments. When these comments were found in the VAX files, they were preserved in this field.
Nuclide	Text	Radionuclide	Specifies H3, U235, U238, or Po.
Data_Comments	Memo	Data review comments	
XLS_Date	Date/time	S_Date in Excel® date format	S_Date and Rev_S_Date are provided in Excel® date format to allow sorting by date and time.
Rev_XLS_Date	Date/time	Rev_S_Date in Excel® date format	
Data_File	Text	VAX data file reference	VAX data file name for original electronic data.
Validated	Yes/no	Logical flag used to indicate validated data record	

LANL staff indicated that a substantial effort had been made to address inconsistencies in the personnel information contained in the VAX files. Particular attention was paid to correcting employee numbers ("Z_Num"), which were not recorded in some of the early laboratory notebooks. LANL attempted to assign Z numbers where possible, but there were approximately 2,000 records with no value in the Z_Num field. Some of the records (311) contained a V number that LANL used at one time for visitors. Other early records (341) contained an alphanumeric identification code.

Minimum detectable concentrations (MDCs) were reported for some, but not all, data. During early LANL operations (before 1970), it was not uncommon for the Laboratory to record only positive bioassay results and not report MDC values. In most cases, "0.00" was reported. The ORAU Team's Access™ database (derived from the primary bioassay database) contains a "MDA" field. The data in this field are a mixture of recording levels and analytical MDA values inferred from procedures or laboratory notes. These values are discussed in the following sections that address the individual nuclides.

Uncertainty estimates were not present in the historical TUPo data. In some cases it was possible to state uncertainties for a specific period based on procedures or laboratory notes. In other cases it was not possible to provide an estimated uncertainty. Uncertainties are discussed for each nuclide in the following sections.

A cursory comparison between the data from the VAX files and that from the laboratory notebooks from which they originated suggested a rather high rate of disagreement (~20%). This prompted an effort to validate a portion of the TUPo data from the VAX files to better gauge its reliability. Approximately 10% of the VAX records were validated against the LANL notebooks from which they originated. This validation process is described in Section 6.0.

The pre-1991 *in vitro* data table in the ORAU Team's database contains a total of 144,624 records for tritium, uranium, and polonium. Table 5-2 shows the number of records and collection date range for each analyte.

Table 5-2. *In vitro* records for tritium, uranium and polonium in the pre-1991 data table.

Analyte	Number of records	Collection date range
H-3	49,218	6/23/1950–12/31/1990
U-235	43,545	4/6/1955–12/31/1990
U-238	46,662	11/18/1949–12/31/1990
Po	5,199	3/11/1947–3/24/1965

The data fields for the pre-1991 *in vitro* data include the "kit type", i.e., the collection protocol; and the sample type. All but 46 of the 144,624 pre-1991 TUPo records are for urinalyses. All but 14 of the urinalyses were spot samples. The remainder were simulated 24 hour collections. The 46 non-urine TUPo records consist of 40 records for spot fecal analyses and 6 for intravenous blood analyses. All 46 of the non-urinalyses records are analyses for polonium.

The analysis type codes for the pre-1991 TUPo records in the ORAU database are shown in Table 5-3. All of the codes are specific to one radioelement or radioisotope.

The T3R analysis type in Table 5-3 includes liquid scintillation counting, used from 1969 forward, and use of an internal GM counter prior to that time (Clark 2005).

Table 5-3. Analysis type codes for pre-1991 TUPo records.

Code	Analysis type
GB	gross alpha/beta (polonium only)
POR	polonium radiometric (polonium only)
T3R	tritium radiometric (tritium only)
UF	uranium fluorophotometric (^{238}U only)
UNAA	delayed neutron activation analysis (^{235}U and ^{238}U only)
UR	uranium radiometric (^{235}U only)

5.2.1 Pre-1991 Tritium Data

50,107 tritium bioassay result records were extracted from the VAX files. These records, covering a period from June 23, 1950 to December 31, 1990, included some analyses for non-LANL workers. These were removed from the ORAU Team's Access™ database. The ORAU database therefore contains 49,218 tritium records. These records cover the same date range as the VAX records, i.e., from June 23, 1950 to December 31, 1990.

Tritium records were extracted from the six VAX data files shown in Table 5-4. Table 5-5 lists the original data fields identified in the six tritium data files.

Table 5-4. Data files with tritium records.

File name	Number of records	Comments
seza5063	17,737	Contains tritium records for 6/23/50 to 12/31/63
seza6475	9,932	Contains tritium records for 1/2/64 to 12/31/74
seza7583	9,961	Contains tritium records for 1/3/75 to 12/30/83
seza8488	8,099	Contains tritium records for 1/3/84 to 12/30/88
st389	1,989	Contains tritium records for 1/3/89 to 12/28/89
st390	2,389	Contains tritium records for 1/2/90 to 12/31/90

Table 5-5. Tritium data file field.

Field	File names		
	seza5063 seza6475 seza7583 seza8488	st389	st390
	Field columns		
Employee Name	2–18	2–18	2–19
Z-Number	19–25	19–24	20–25
Employee Code	26–31	25–34	26–31
Birth Year	32–33	Not used	32–33
Sample Date	36–41	35–40	36–41
Sample Type Code	Not used	41–42	Not used
Results	42–51	43–50	42–51
LANL Notebook ID	Not used	51–57	Not used
LANL Notebook Page	Not used	58–60	Not used
Analyst ID	Not used	61–66	Not used
Analysis Type Code	Not used	67–69	Not used
Sequential Record ID	Not used	70–75	Not used

5.2.1.1 Tritium Reporting Units

In the VAX files, all tritium results were reported using the units code "UL", indicating microcuries per liter. Tritium results were never corrected for void time. Because historical documentation indicated that all tritium results were calculated in terms of microcuries per liter, any results for which units were

missing were updated to reflect these units. Thus, all of the tritium results in the ORAU database are in units of $\mu\text{Ci/L}$.

5.2.1.2 Tritium Sample Type Codes

Tritium sample type codes were not always recorded in the VAX files. However, those records for which a sample type code was given always showed "U1", indicating a spot urine sample. Thus, all of the tritium records in the ORAU database are spot urine samples.

5.2.1.3 Tritium Analysis Type Codes

The tritium VAX files contained the analysis type codes "H3R", "T3R", "33", "H3", "T#R", and "T3". There were 242 records for which no analysis type code was entered. Although two different analytical techniques were used for tritium analysis (Clark 2005), it appears that codes "H3R" and "T3R" were used interchangeably and did not distinguish between the two different techniques. Thus, the analysis type codes "H3R", "33", "H3", "T#R", and "T3" were all changed to T3R for consistency. All of the tritium records in the ORAU database therefore show T3R as the analysis type code.

5.2.1.4 Tritium MDCs

The recording of MDCs in the VAX files varied over time. Early data contained no MDC information, with nondetections being reported as "0.00." Later data indicated MDC values by using an "L" as a prefix (e.g., "L1.00" meant less than 1.00). To present MDC information consistently in the new database, all results below MDC were reported as "0.00" in the sample result field.

Unlike the other TUPo records, all of the tritium records in the ORAU database have a value in the "MDA" field. ("MDA" is the name of the field in the ORAU database that contains MDC values.) These values are shown in Table 5-6. These MDC values should be regarded as recording levels.

Table 5-6. MDC values for tritium bioassay records in the ORAU database.

Period	MDC Value
6/23/1950–12/9/1987	1.0 $\mu\text{Ci/L}$
12/11/1987–12/31/1990	0.1 $\mu\text{Ci/L}$

5.2.1.5 Tritium Uncertainty

Uncertainty information was not recorded in the VAX files. Reviews of laboratory procedures, notebooks, and memoranda provided some information for uncertainty estimates. This information is summarized in Table 5-7.

Table 5-7. Uncertainties for tritium bioassay records.

Period	Analytical method	Uncertainty statement	Reference
ca. 1954	Internal Geiger-Müller	$\pm 5\%$ between the range of 1 to 250 $\mu\text{Ci/L}$	McClelland et al. (1955, Ch. 26, p. 140)
1958–1967	Internal Geiger-Müller	1 to 2 $\mu\text{Ci/L}$ at 1 $\mu\text{Ci/L}$ 9.5 to 10.5 $\mu\text{Ci/L}$ at 10 $\mu\text{Ci/L}$ 99 to 101 $\mu\text{Ci/L}$ at 100 $\mu\text{Ci/L}$	Lawrence (1958)
1968–1990	Liquid scintillation	$\pm 5\%$ (1-sigma) at 1 $\mu\text{Ci/L}$	Lawrence (1987)

5.2.2 Pre-1991 Uranium Data

Uranium bioassay records were extracted from the VAX data files listed in Table 5-8 and 5-9 for " ^{235}U " and " ^{238}U ", respectively. It is important to note that LANL used " ^{235}U " to refer to enriched uranium and

²³⁸U” to refer to depleted or normal uranium. The use of ²³⁵U” or ²³⁸U” in bioassay results did not imply that radionuclide-specific analyses were performed.

Table 5-8. Data files with ²³⁵U” records.

File name	Records	Comments
sag5574	8,953	Contains U-235 records for last names A to G for 4/08/55 to 12/20/74.
shn5574	7,352	Contains U-235 records for last names H to N for 4/11/55 to 12/20/74.
soz5574	10,151	Contains U-235 records for last names O to Z for 4/06/55 to 12/20/74
su57581	11,594	Contains U-235 records for 1/03/75 to 12/25/81.
su235a-z.1982-88	4,929	Contains U-235 records for dates 12/25/81 to 2/19/88. Also includes two records dated 7/09/62 that are of questionable value based on examination of notebook data.
su5o89	272	Contains U-235 records for 1/03/89 to 12/18/89
su5o90	400	Contains U-235 records for 1/02/90 to 12/31/90

Table 5-9. Data files with ²³⁸U” records.

File name	Number of records	Comments
su238a	9,889	Contains U-238 records for last names A to G for 11/18/49 to 12/13/74.
su238b	9,699	Contains U-238 records for last names H to N for 11/18/49 to 12/13/74.
su238c	10,014	Contains U-238 records for last names O to Z for 11/18/49 to 12/13/74.
su87581	11,830	Contains U-238 records for 1/03/75 to 12/25/81.
su238a-z.1982-88	5,159	Contains U-238 records for 2/19/81 to 12/21/88.
su8o89	604	Contains U-238 records for 1/03/89 to 12/19/89.
su8o90	959	Contains U-238 records for 1/02/90 to 12/31/90.
sPOL4765.dat	6	These records were found in the polonium data files.

The original data fields identified in the uranium data files are listed in Tables 5-10 and 5-11 for the ²³⁵U” and ²³⁸U” records, respectively.

Table 5-10. ²³⁵U” data file fields.

Field	File names		
	sag5574 shn5574 soz5574	su57581 su235a-z.1982-88	su5o89 su5o90
	Field columns		
Employee Name	2–19	2–19	1–18
Z-Number	20–24	20–24	19–23
Employer Code	25–34	25–34	24–33
Sample Date	35–40	35–40	34–39
Sample Type Code	41–42	41–42	Not used
Results	43–50	43–50	40–47
Result Units	51–52	51–52	48–49
LANL Notebook ID	53–57	53–57	Not used
LANL Notebook Page	58–60	58–60	Not used
Analyst ID	61–67	61–67	Not used
Analysis Type Code	68–69	68–72	52–55
Nuclide ID	Not used	73–76	57–60

Table 5-11. ^{238}U data file fields.

Field	File names		
	su238a su238b su238c	su87581 su238a-z.1982-88	su8o89 su8o90
	Field columns		
Employee Name	2–19	2–19	1–18
Z-Number	20–24	20–24	19–23
Employer Code	25–34	25–34	24–33
Sample Date	35–40	35–40	34–39
Sample Type Code	41–42	41–42	Not used
Results	43–50	43–50	40–47
Result Units	51–52	51–52	48–49
LANL Notebook ID	53–57	53–57	Not used
LANL Notebook Page	58–60	58–60	Not used
Analyst ID	61–67	61–67	Not used
Analysis Type Code	68–69	68–72	52–55
Notebook Comments	70–85	Not used	Not used
Nuclide ID	86–90	73–76	57–60
Sequential Record ID	93–97	Not used	Not used

5.2.2.1 Uranium Reporting Units

From 1955 through 1976 urinalysis results for the ^{235}U records in the VAX files were reported with a unit code of “DM” for disintegrations per minute per liter. Starting in 1977, the ^{235}U unit code became “PL” for picocuries per liter. ^{238}U bioassay results in the VAX files were reported using the unit code “UL” for micrograms per liter. In the ORAU database, all of the ^{235}U results are reported in units of picocuries per liter. All ^{238}U results are reported in micrograms per liter.

5.2.2.2 Uranium Sample Type Codes

The uranium sample type codes found in the VAX files were “U1”, “U4”, “U5”, “U6”, and “U8”. One record indicated a sample type code of “U1Y”, which was assumed to be an error and changed to “U1”.

The “U1” sample type code specified a spot urine sample. Sample type code “U5” indicated a four-bottle kit that was collected as a true 24-hour collection. The code “U6” was only used for 2 ^{235}U results and 20 ^{238}U results.

In the ORAU database, all but 14 of the ^{235}U records are spot urine samples. The other 14 records are simulated 24 hour urine samples collected in August and September of 1969. All of the ^{238}U records are spot urine samples.

5.2.2.3 Uranium Analysis Type Codes

The uranium analysis type codes found in the VAX files are listed in Table 5-12. Of these, only three (“UF”, “UNAA”, and “UR”) appeared to reflect analysis types that were routinely used in a given era. The remaining analysis type codes were treated as ad hoc designations or transcription errors because of the few times they occurred. These were changed to UF, UNAA, or UR, as appropriate, based on analyte and analysis date. Uranium neutron activation analysis (UNAA) is discussed in Attachment A.

In the ORAU database, all of the ^{235}U records have an analysis type of either UR or UNAA. All of the ^{238}U records are either UF or UNAA.

Table 5-12. Uranium analysis type codes.

Analysis code	Meaning	Record type
46	Meaning unknown—referenced once	U-238
COLOR	Meaning unknown—referenced 3 times	U-238
CPMS	Meaning unknown—referenced once	U-238
U	Meaning unknown—referenced 7 times	U-238
U6	Meaning unknown—referenced once	U-238
U6F	Meaning unknown—referenced once	U-238
U9	Meaning unknown—referenced 5 times	U-235
UF	Uranium fluorophotometric	U-238
UNAA	Delayed Neutron Activation Analysis	U-235 and U-238
UR	Uranium radiometric	U-235

5.2.2.4 Uranium MDCs

MDC information was not provided in the VAX data until 1962 for “²³⁵U” records and until 1968 for “²³⁸U” records. The values in the “MDA” field of the ORAU database for the ²³⁵U and ²³⁸U records are shown in Table 5-13. Values are given by date range for the two analysis type codes used for each nuclide, and include information on records for which no MDC value is provided. For these records the MDC value is given as “null.” For the ²³⁵U records, all of the MDC values are in units of picocuries per liter. In cases where the “MDA” value in the database reflects a units conversion from its historical value, the historical value is shown in parentheses. In general the MDC values in the ORAU database, where given, should be regarded as recording levels.

Table 5-13. MDC values for uranium bioassay records in the ORAU database.

Nuclide	Analysis type	Period	“MDA” value
U-235	UR (uranium radiometric)	4/6/1955–2/6/1981	null
		7/31/1962–1/17/1964	3.15 pCi/L (7 dpm/L)
		1/7/1977–11/24/1980	4.00 pCi/L
		6/18/1976–12/17/1976	4.50 pCi/L (10 dpm/L)
		3/19/1971–6/4/1976	5.41 pCi/L (12 dpm/L)
		11/8, 11/12, & 11/22 1963 (3 records)	6.76 pCi/L (15 dpm/L)
		12/20/1974 (1 record)	7.00 pCi/L
		UNAA (delayed neutron activation analysis)	8/19/1977–8/29/1988*
	8/12/1977–12/31/1990*	4.00 pCi/L	
U-238	UF (uranium fluorophotometric)	11/18/1949–8/20/1982	null
		3/1/1976–8/25/1978	1.00 µg/L
		1/15/1968–8/20/1982	4.00 µg/L
		9/3/1982 (1 record)	20.00 µg/L
	UNAA (delayed neutron activation analysis)	1/13/1978–12/19/1988	null
		1/13/1978–6/25/1982	1.00 µg/L
		7/6/1982–12/31/1990	4.00 µg/L
		1/7/1983 (23 records)	11.00 µg/L

*There are two ²³⁵U records that show an analysis type of UNAA and a sample collection date of 7/9/1962. These are assumed to be an error either in the collection date or the analysis type.

5.2.2.5 Uranium Uncertainty

Uncertainty information was not recorded in the VAX data files. Reviews of laboratory procedures, notebooks, and memoranda provided the information in Table 5-14.

Table 5-14. Uncertainties for uranium bioassay records.

Record type	Period	Analytical method	Uncertainty statement	Reference
U-235	Uncertain	Radiometric	At spike levels of 32 and 64 dpm/L recovery is 83% ±16%	LANL (1978, Section 13)
	Starting 1978	Delayed neutron counting	±0.4 pCi/L at 5.2 pCi/L ±0.6 pCi/L at 13.9 pCi/L ±1.6 pCi/L at 26.1 pCi/L overall: ±6% (1 sigma)	Gautier (1983); LANL (1983)
U-238	1949–1955 and perhaps up to 1978	Fluorophotometric	±10%	McClelland et al. (1955, Ch. 27, p. 155)
	In place in 1978 but possibly earlier	Fluorophotometric	±4% relative standard deviation at 20 µg/L	LANL (1978, Section 14)
	Procedure effective date 10/1/82	Colorimetric	Average recovery of standard is ±8% at the 11 µg/50 cm ³ concentration level	Gautier (1983); LANL (1983)
	Starting 1978	Delayed neutron counting	±0.3 µg/L at 4.2 µg/L ±0.4 µg/L at 8.5 µg/L ±0.5 µg/L at 16.9 µg/L Overall: ±8% (1 sigma)	Gautier (1983); LANL (1983)

5.2.3 Pre-1991 Polonium Data

Bioassay records for polonium were extracted from the VAX data files shown in Table 5-15. The original data fields identified for the polonium data files are given in Table 5-16.

Table 5-15. Polonium records data file.

File name	No. of records	Comments
sPOL4765.dat	5,676	3/11/47 to 3/24/65

Table 5-16. Polonium data file fields.

Field	File name
	SPOL4765.dat
	Field columns
Employee Name	2–19
Z-Number	20–24
Employer Code	25–34
Sample Date	35–40
Sample Type Code	41–42
Results	43–50
Result Units	51–52
LANL Notebook ID	53–57
LANL Notebook Page	58–60
Analyst ID	61–66
Analysis Type Code	67–69

5.2.3.1 Polonium Reporting Units and Unit Conversion

The polonium bioassay results in the VAX files were initially reported with a unit code of “CM” for counts per minute per 24 hours. Starting in November of 1952 results began to be reported using a unit code of “DM” for disintegrations per minute per liter. A few results continued to be reported as “CM” through January of 1953. For consistency, the “DM” data were converted to picocuries per day by assuming a urine output of 1.4 L/d as follows:

$$dpm/L \times 0.63 = pCi/d \quad (1)$$

Counts per minute data were not converted because the counting efficiencies were unknown.

In addition to routine urinalysis results, the polonium data included records for 6 blood and 40 fecal analyses. Blood analysis results were reported using the unit code “DM”, which in this case indicated disintegrations per minute per milliliter of blood. Fecal results were reported using the unit code “NCI” for nanocuries per gram.

All of the polonium records in the ORAU database, including those for blood and fecal analyses, have results in units of either cpm per liter or picocuries per liter. The date ranges for these overlap: results in cpm/L occur from 3/11/1947 through 1/9/1953, and those in pCi/L occur from 2/10/1948 through 3/24/1965.

5.2.3.2 Polonium Sample Type Codes

Four sample type codes were present in the polonium data extracted from the VAX files: “U1”, “U6”, “B1”, and “F7”. The first two indicated urine samples, with “U1” signaling a spot sample and “U6” specifying a “simulated” 24-hour sample. Sample type code “B1” was used for blood samples, and “F7” was used for fecal samples.

Of the 5,199 polonium records in the ORAU database, 40 are spot fecal samples, 6 are intravenous blood samples, and the remainder are spot urine samples.

5.2.3.3 Polonium Analysis Type Codes

The polonium VAX files contained the analysis type codes “GB” or “POR”. “GB” indicated gross alpha and beta and was used for only 13 polonium records. “POR” indicated polonium radiometric analysis.

In the ORAU database, there are the 13 GB analyses and 5,186 POR results. The 13 GB analyses were performed for spot urine samples collected on 2/18/1960 (1 record), 2/19/1960 (8 records) , or 2/23/1960 (10 records). The POR analyses were performed for spot urine, spot fecal, or intravenous blood samples over the period 3/11/1947 through 3/24/1965.

5.2.3.4 Polonium MDCs and Uncertainty

The polonium VAX files contained no information on MDCs or uncertainty. MDCs were inferred from procedures and reports. Gautier (1983, Procedure R160) describes the general method for polonium bioassay analysis and provides the MDC and uncertainty values given in Tables 5-17 and 5-18.

Table 5-17. MDCs for polonium bioassay records.

Period	Analytical method	Analytical MDC
Up to 1955	Radiometric	10 dpm/L
1955–1965	Radiometric	0.1 pCi/L

Table 5-18. Polonium uncertainty.

Period	Analytical method	Uncertainty statement	Reference
Effective 5/1/1955	Radiometric	±3% (±1 sigma) at the 15-pCi concentration level	Gautier (1983, Procedure R160-1)

In the ORAU database, none of the 13 polonium records analyzed via gross alpha/beta counting (analysis type GB) have a MDC reported in the “MDA” field. The MDC values given for the 5,186 records analyzed by polonium radiometric analysis are given in Table 5-19.

Table 5-19. MDC values for polonium bioassay records in the ORAU database.

Period	Analysis Type	"MDA" Value
3/11/1947–3/24/1965	POR	null
1/3/1955–12/14/1955	POR	0.1 pCi/L
1/11/1954–8/10/1955	POR	4.50 pCi/L (10 dpm/L)

5.3 POST-1991 *IN VITRO* DATA

The post-1991 *in vitro* data table in the ORAU Team's database contains a total of 244,760 records covering a range of collection dates from 12/29/1973 to 2/9/2009. Table 5-20 shows the analytes, numbers of records, and collection dates for the post-1991 *in vitro* data. In many cases there are multiple records for a given bioassay sample, as some samples were subjected to more than one type of analysis. This is particularly true for plutonium analyses. Thus, the number of records exceeds the actual number of bioassays. For each record the analysis result (if any) is reported in one of three ways: activity in terms of normalized excretion or excretion rate, activity per sample, or activity per aliquot. Only one of the three result types is reported for a given record, though more than one result may be reported for a given sample and analyte. This is seen with the plutonium and americium analyses, where samples may be analyzed by more than one analysis technique depending on what the initial count showed. In these cases multiple analysis results can appear for the same sample and analyte, with the results reported either as the same type or on different bases (e.g., on a normalized excretion basis and per sample basis). There are also records for which no result of any type is reported. This occurs with nearly all of the *in vitro* analytes, but is especially prevalent for the ^{240}Pu analyses, where approximately half of the records do not show a result. These null results for ^{240}Pu reflect the use of mass spectrometry analysis. It is not clear why there appears to be a gap in the *in vitro* bioassay records for January through June of 1991. The gap exists for the ^{241}Am and $^{238,239}\text{Pu}$ analyses in addition to tritium and uranium.

Table 5-20. *In vitro* records in the post-1991 table of the ORAU database.

Analyte	Urinalysis records	Fecal analysis records	Total records	Collection date range	
				From	To
AM-241	2,456	201	2,657	12/29/1973	1/19/2009
H-3	28,246	0	28,246	6/17/1991	2/9/2009
PO-210	69	0	69	6/13/2007	10/3/2008
PU-238	57,000	252	57,252	12/29/1973	1/30/2009
PU-239	77,351	252	77,603	12/29/1973	1/30/2009
PU-239+PU-240	13,517	0	13,517	12/6/1996	1/30/2009
PU-240	12,487	0	12,487	1/26/1997	1/30/2009
TH-228	13	0	13	1/27/2005	5/28/2008
TH-229	13	0	13	1/27/2005	5/28/2008
TH-232	4	0	4	1/20/2008	5/28/2008
U-234	17,627	6	17,633	7/1/1991	1/21/2009
U-235	17,627	6	17,633	7/1/1991	1/21/2009
U-238	17,627	6	17,633	7/1/1991	1/21/2009

The post-1991 *in vitro* dataset received from LANL contained a number of duplicate records. Duplicates in this context means the same result being reported more than once for the same Sample ID. It does not refer to cases where there are different results reported for the same sample, reflecting different analysis types or reporting basis.

The number of duplicate records noted for each *in vitro* analyte are summarized in Table 5-21 for each of the three types of analysis result. Activity_PA and Activity_PS refer to the activity per analysis and per sample, respectively. Activity refers to activity in terms of excretion. These are the field headings used in the *in vitro* data table. An entry of n/a in Table 5-21 means there were no results of

that type in the dataset for that analyte. The numbers of duplicates are the same for the three uranium isotopes because all uranium samples are analyzed for all three nuclides. Likewise, there is a ^{239}Pu result corresponding to each ^{238}Pu result, though there can be additional ^{239}Pu results for a given sample if follow-up analysis was performed.

Table 5-21. Duplicate records in the post-1991 *in vitro* data table of the ORAU database.

Analyte	Number of duplicate records		
	Activity_PA	Activity_PS	Activity
AM-241	56	1	67
H-3	n/a	n/a	28
PO-210	0	n/a	n/a
PU-238	0	0	2
PU-239	0	0	2
PU-239+PU-240	n/a	0	n/a
PU-240	n/a	0	0
TH-228	0	n/a	n/a
TH-229	0	n/a	n/a
TH-232	0	n/a	n/a
U-234	329	51	834
U-235	329	51	834
U-238	329	51	834

For all analytes except plutonium, the duplicate records were cases where the same results were reported with the same sample volume given in different units. Specifically, the duplicates occurred when the same result was reported twice: once with the sample volume stated in grams, and a second time with the same sample volume stated in volume units (liters or milliliters). For plutonium, there are only two samples (4 records) that were duplicated. These were cases where the same results were reported twice for the same samples, but showed different analysis types. Users should be aware of the possibility of duplicate records in the post-1991 *in vitro* dataset and confirm that there is not more than one instance of the same result for a given Sample ID.

The data fields for the post-1991 *in vitro* data include the schedule type and the kit type. There are three possible values for the schedule type and six different kit types. The three schedule types and the corresponding fraction of the total post-1991 *in vitro* records are Prompt Action (1%), Special (14%) and Routine (85%). Thus, the post-1991 *in vitro* data are predominantly routine bioassays.

The six kit types are 500 mL Sample, Simulated 24 Hour, Spot Fecal, Spot Sample, Timed Voidings, and True 24 Hour. The 500 mL Sample kit was used for uranium, ^{210}Po , ^{228}Th (1 record), and ^{229}Th (1 record). The Spot Sample was used only for tritium.

Table 5-22 shows the breakdown of the post-1991 *in vitro* records by kit type for each of the three schedule types.

Table 5-22. Breakdown of kit types for post-1991 *in vitro* records.

Schedule type	Kit type	Fraction
Prompt action	500 ml sample	1%
	Simulated 24 hour	79%
	Spot fecal	9%
	Spot sample (tritium only)	1%
	Timed voidings	--
	True 24 hour	10%
Special	500 ml sample	5%
	Simulated 24 hour	89%

Schedule type	Kit type	Fraction
	Spot fecal	0.2%
	Spot sample (tritium only)	5%
	Timed voidings	0.3%
	True 24 hour	1%
Routine	500 ml sample	25%
	Simulated 24 hour	63%
	Spot fecal	--
	Spot sample	13%
	Timed voidings	--
	True 24 hour	0% (13 records)

The Timed Voidings kit type was only associated with Special analyses. Spot fecal collections were not associated with any of the Routine sampling records.

The analysis type codes seen in the post-1991 *in vitro* data are shown in Table 5-23. The PHW analysis type was only used for fecal samples, though not all fecal samples were counted by PHW. Most were counted by RAS. TIMS analysis is only used for plutonium analyses.

Table 5-23. Analysis type codes for post-1991 *in vitro* records.

Code	Analysis type
LS	Liquid scintillation counting (tritium only)
PHW	Phoswich detector
RAS	Radiometric alpha spectrometry
RASLC	Radiometric alpha spectrometry (unknown variant)
RASRC	Radiometric alpha spectrometry (unknown variant)
RASSP	Radiometric alpha spectrometry (unknown variant)
TIMS	Thermal ionization mass spectrometry

5.3.1 Post-1991 In Vitro Data for Tritium

The post-1991 *in vitro* data table in the ORAU Team's database contains 28,246 records for tritium analyses. 26,667 (94%) of these were routine collections over the period 6/17/1991 to 2/9/2009. 16 of the tritium records were from prompt action collections over a three day span in August of 1996. The remaining 1,563 (6%) were special collections over the period 7/1/1991 to 1/8/2009. All tritium records show a spot sample collection protocol and analysis by liquid scintillation counting (analysis type code LS).

All but two of the 26,667 routine urinalysis records for tritium show a non-null result. All of the non-null results are reported in terms of excretion, i.e. none are on a per sample or per aliquot basis. Most of the results are reported in units of μCi per mL. 2 are reported in μCi per gram and 37 do not have units. 2,289 of the results are negative and 2,993 are zero.

16,324 (61%) of the routine tritium urinalysis records have a MDC reported, though 5 of these show a MDC of zero. The units for the MDC values are primarily $\mu\text{Ci/L}$. 32 records give a MDC value in pCi/L and 2 are in $\mu\text{Ci/gram}$. Records that have a MDC reported include those where the results are negative or zero. The units for the MDC can sometimes differ from that for the corresponding result.

5.3.2 Post-1991 In Vitro Data for Polonium

The post-1991 *in vitro* data table contains 69 records for ^{210}Po urinalyses. These samples were collected over a date range of 6/13/2007 to 10/3/2008. Two of them were denoted routine samples and the rest were special collections. The two routine samples were simulated 24 hour urine

samples. Of the 67 special collections, 2 were simulated 24 hour samples and the rest were 500 mL samples.

A non-null result is reported for all of the ^{210}Po samples. All of the results are on a per aliquot basis. There is one negative result, and no zero results. All results have a corresponding MDC given in pCi/aliquot. Sample volumes are given for all results, in grams.

5.3.3 Post-1991 In Vitro Data for Thorium

The post-1991 *in vitro* data table contains 13 records for ^{228}Th , 13 records for ^{229}Th , and 4 records for ^{232}Th . These 30 records correspond to 13 urinalysis samples: 9 analyzed for ^{228}Th and ^{229}Th only, and 4 analyzed for all three thorium isotopes. The analyses for ^{228}Th and ^{229}Th only occurred between 1/27/2005 and 1/23/2008. The analyses for all three isotopes occurred in January (3 samples) and May (1 sample) of 2008. All analyses were by RAS. All 13 samples were special collections. Twelve (12) were simulated 24 hour samples and the other was a 500 mL sample. Results are reported for all 30 records in pCi per aliquot. All 30 records report MDCs, also in pCi per aliquot, and sample size in grams.

5.3.4 Post-1991 In Vitro Data for Uranium

The post-1991 *in vitro* data table contains 52,899 records for uranium analyses, i.e., analyses for ^{234}U , ^{235}U , or ^{238}U . These records represent 17,633 samples, as each sample was analyzed for the three uranium isotopes. Overall the uranium samples cover a date range of 7/1/1991 to 1/21/2009. Table 5-24 shows a summary of the post-1991 uranium records.

Table 5-24. Summary for uranium records in the post-1991 *in vitro* data.

Schedule type (no. records)	Kit type (no. records)	Collection date range
Prompt action (42)	500 mL sample (24)	10/5/1994–10/29/1998
	Spot fecal (18)	3/7/2005–3/10/2005
Special (1,731)	500 mL sample (1,716)	8/2/1991–5/26/2008
	Simulated 24 hour (15)	1/27/2005–2/1/2005
Routine (51,126)	500 mL sample (51,126)	7/1/1991–1/21/2009

The analysis type for all but 9 of the uranium records (3 samples) was RAS. The other 9 records showed an analysis type of RASSP. These 3 samples were special collections of 500 mL samples collected on 12/17/2007 and 12/18/2007.

All but three of the 51,126 routine sample records (representing 17,042 uranium urinalyses) have a non-null result in the database. Thus, a result is given for all but 1 of the 17,042 samples. Results are reported for each of the three result types, as shown in Table 5-25.

Table 5-25. Non-null results for routine uranium analyses in the post-1991 *in vitro* data.

Result type	No. non-null records	No. records less than zero	No. records = zero
Per aliquot	15,264 (5,088 samples)	1,792	459
Per sample	306 (102 samples)	34	12
Excretion basis (pCi/L)	35,553 (11,851 samples)	7,494	1,942

MDC values are given for 10,341 (29%) of the 35,553 non-null routine sample results reported in terms of excretion (pCi/L). The units for these are primarily in pCi/aliquot (48%) and pCi/sample (50%). Less than 2% are in consistent units with the excretion result. All of the per aliquot and per

sample results have a MDC reported. All of these are in consistent units, though the MDC for 3 of the per aliquot results is zero.

Sample volumes are given for all of the per aliquot and per sample results, and for all but 3 of the excretion results. For the per aliquot and per sample results the sample volumes are given in grams or mL. For the excretion results they are given in grams or liters.

5.3.5 Post-1991 In Vitro Data for Plutonium

There are 4 individual analytes in the post-1991 *in vitro* data for plutonium: ^{238}Pu , ^{239}Pu , ^{240}Pu , and $^{239}\text{Pu}+^{240}\text{Pu}$. Overall, the post-1991 *in vitro* data table contains a total of 160,859 records for plutonium analyses covering a range of sample collection dates from 12/29/1973 to 1/30/2009. 80% of the plutonium records were routine analyses, 19% were special, and 2% were prompt action.

Plutonium analyses account for two thirds of the post-1991 *in vitro* bioassay records. This is misleading, however, as a single plutonium sample can have up to 9 associated records (results). For a given plutonium sample, results may be reported reflecting different bases (e.g. excretion and per sample bases), different analysis techniques, and additional analytes; depending on the results from the initial analysis. Thus, the 160,859 plutonium records in the post-1991 *in vitro* dataset represent 39,181 individual samples. Table 5-26 shows the distribution of plutonium records associated with individual plutonium samples. Each plutonium sample has at least two associated records since at a minimum each one is assayed for ^{238}Pu and ^{239}Pu . These two results become four when they are reported in terms of both activity per sample and on an excretion rate basis. Additional results are created for the same sample if a follow-up TIMS analysis is performed, which allows ^{239}Pu and ^{240}Pu to be differentiated. Results for these additional analytes may also be reported on both a per sample and excretion rate basis.

Table 5-26. Distribution of plutonium records per sample in the post-1991 *in vitro* data.

No. plutonium records per sample	Number of samples
2	14,681
3	1
4	13,233
5	4,174
6	95
7	2,855
8	141
9	4,001

71% of the plutonium analyses were performed via RAS. These include a small number of records analyzed via the RASLC (16 records) and RASRC (12 records) analysis types. A small number (172 out of 160,859) of analyses were performed using a phoswich detector (analysis type PHW). These were all fecal analyses, and each has a corresponding RAS count (i.e. there is a PHW count and a RAS count for each fecal analysis). The remainder (29%) were analyzed via TIMS, which LANL began using in late 1996. TIMS analysis allows ^{239}Pu and ^{240}Pu activity to be determined independently. TIMS analysis can be associated with all plutonium analytes except ^{238}Pu . All $^{239}\text{Pu}+^{240}\text{Pu}$ and ^{240}Pu results are via TIMS analysis.

5.3.5.1 Post-1991 In Vitro Data for ^{238}Pu

The post-1991 *in vitro* data table contains 57,252 records for ^{238}Pu analyses. These are summarized in Table 5-27.

Table 5-27. Summary for ^{238}Pu records in the post-1991 *in vitro* data.

Collection requirement	Collection protocol (no. records)	Collection date range	Analysis types (no. records)
Prompt action (1,148 records)	Simulated 24 hour (1,010)	2/12/1992–11/23/2008	RAS (935), PHW (73), RASLC (1), RASRC (1)
	Spot fecal (43)	8/8/1989–3/10/2005	RAS (39), PHW (4)
	True 24 hour (95)	1/21/1993–1/24/2007	RAS (88), PHW (7)
Special (10,587 records)	Simulated 24 hour (10,426)	7/1/1991–1/30/2009	RAS (10,417), RASLC (6), RASRC (3)
	Spot fecal (32)	12/29/1973–6/2/2006	RAS (30), PHW (2)
	Timed voidings (23)	4/1/1992–4/3/2000	RAS (23)
	True 24 hour (106)	9/16/1991–2/11/2008	RAS (106)
Routine (45,517 records)	Simulated 24 hour (45,514)	4/8/1991–1/12/2009	RAS (45,511), RASLC (1), RASRC (2)
	True 24 hour (3)	5/28/2003–6/22/2005	RAS (3)

All but 35 of the 45,517 routine urinalysis records for ^{238}Pu have a non-null result in the database. Results are reported for two of the three result types, as shown in Table 5-28. There are no routine analysis results for ^{238}Pu reported on a per aliquot basis.

Table 5-28. Non-null results for routine ^{238}Pu analyses in the post-1991 *in vitro* data.

Result type	No. non-null records	No. records less than zero	No. records = zero
Per aliquot	0	n/a	n/a
Per sample	21,572	9,997	965
Excretion basis (pCi/24 hrs)	23,910	11,429	1,888

Sample volumes are reported for 60% of the non-null excretion results and for all but 107 of the per sample results. All sample volumes are given in grams. 61% of the excretion results have a MDC reported. Two of these MDC values are zero and 10 are negative. All are in units of pCi per sample. For the per sample results, all but 2 have a MDC reported. Two of these MDC values are zero and 16 are negative. The units are also pCi per sample.

5.3.5.2 Post-1991 *In Vitro* Data for ^{239}Pu

The post-1991 *in vitro* data table contains 77,603 records for ^{239}Pu analyses. These are summarized in Table 5-29.

Table 5-29. Summary for ^{239}Pu records in the post-1991 *in vitro* data.

Collection requirement	Collection protocol (no. records)	Collection date range	Analysis types (no. records)
Prompt action (1,290 records)	Simulated 24 hour (1,102)	2/12/1992–11/23/2008	RAS (935), TIMS (92), PHW (73), RASLC (1), RASRC (1)
	Spot fecal (46)	8/8/1989–3/10/2005	RAS (39), PHW (4), TIMS (3)
	True 24 hour (142)	1/21/1993–1/24/2007	RAS (88), TIMS (47), PHW (7)
Special (14,536 records)	Simulated 24 hour (14,318)	7/1/1991–1/30/2009	RAS (10,417), TIMS (3,892), RASLC (6), RASRC (3)
	Spot fecal (32)	12/29/1973–6/2/2006	RAS (30), PHW (2)
	Timed voidings (42)	4/1/1992–4/3/2000	RAS (23), TIMS (19)
	True 24 hour (144)	9/16/1991–2/11/2008	RAS (106), TIMS (38)
Routine (61,777 records)	Simulated 24 hour (61,771)	4/8/1991–1/12/2009	RAS (45,511), TIMS (16,257), RASRC (2), RASLC (1)
	True 24 hour (6)	5/28/2003–6/22/2005	RAS (3), TIMS (3)

All but 143 of the 61,777 routine urinalysis records for ^{239}Pu have a non-null result in the database. Results are reported for two of the three result types, as shown in Table 5-30. There are no routine analysis results for ^{239}Pu reported on a per aliquot basis. 30 of the excretion results are reported in units of pCi per sample. The remainder are in pCi per 24 hours.

Table 5-30. Non-null results for routine ^{239}Pu analyses in the post-1991 *in vitro* data.

Result type	No. non-null records	No. records less than zero	No. records = zero
Per aliquot	0	n/a	n/a
Per sample	32,245	13,098	1,006
Excretion basis	29,389	11,832	2,283

Sample volumes are reported for 51% of the non-null excretion results and for 68% of the per sample results. All sample volumes are given in grams. 49% of the excretion results have a MDC reported. Two of these MDC values are zero and 10 are negative. All are in units of pCi per sample. For the per sample results, 67% have a MDC reported. Two of these MDC values are zero and 16 are negative. The units are also pCi per sample.

5.3.5.3 Post-1991 *In Vitro* Data for ^{240}Pu

The post-1991 *in vitro* data table contains 12,487 records for ^{240}Pu analyses. These are summarized in Table 5-31. The analysis type for all ^{240}Pu records is TIMS.

Table 5-31. Summary for ^{240}Pu records in the post-1991 *in vitro* data.

Collection requirement	Collection protocol (no. records)	Collection date range
Prompt action (79 records)	Simulated 24 hour (57)	7/23/1998–11/23/2008
	Spot fecal (1)	3/6/1998
	True 24 hour (21)	6/11/1997–3/27/2001
Special (2,307 records)	Simulated 24 hour (2,267)	2/13/1997–1/30/2009
	Timed voidings (15)	5/5/1997–6/26/1997
	True 24 hour (25)	1/26/1997–2/11/2008
Routine (10,101 records)	Simulated 24 hour (10,100)	2/10/1997–12/24/2008
	True 24 hour (1)	6/9/2004

4,905 (49%) of the 10,101 routine urinalysis records for ^{240}Pu have a non-null result in the database. Results are reported for two of the three result types, as shown in Table 5-32. There are no routine analysis results for ^{240}Pu reported on a per aliquot basis. All of the 703 results in terms of excretion have a corresponding result on a per sample basis.

Table 5-32. Non-null results for routine ^{240}Pu analyses in the post-1991 *in vitro* data.

Result type	No. non-null records	No. records less than zero	No. records = zero
Per aliquot	0	n/a	n/a
Per sample	4,202	1,659	1
Excretion basis (pCi/24 hrs)	703	148	1

Sample volumes, in grams, are reported for 33 records for both the per sample results and the excretion results. MDC values are not provided for TIMS analyses.

5.3.5.4 Post-1991 *In Vitro* Data for $^{239}\text{Pu}+^{240}\text{Pu}$

The post-1991 *in vitro* data table contains 13,517 records for $^{239}\text{Pu}+^{240}\text{Pu}$ analyses. These are summarized in Table 5-33. The analysis type for all $^{239}\text{Pu}+^{240}\text{Pu}$ records is TIMS.

Table 5-33. Summary for $^{239}\text{Pu}+^{240}\text{Pu}$ records in the post-1991 *in vitro* data.

Collection requirement	Collection protocol (no. records)	Collection date range
Prompt action (99 records)	Simulated 24 hour (53)	12/6/1996–11/23/2008
	Spot fecal (3)	3/6/1998, 3/10/1998, 3/11/1998
	True 24 hour (43)	12/20/1996–1/20/2007
Special (2,596 records)	Simulated 24 hour (2,539)	12/22/1996–1/30/2009
	Timed voidings (19)	5/5/1997–6/27/1997
	True 24 hour (38)	12/20/1996–2/11/2008
Routine (10,822 records)	Simulated 24 hour (10,819)	2/6/1997–12/24/2008
	True 24 hour (3)	5/28/2003, 6/9/2004, 6/22/2005

10,714 (99%) of the 10,822 routine urinalysis records for $^{239}\text{Pu}+^{240}\text{Pu}$ have a non-null result in the database. All of the results are reported on a per sample basis. 2,485 of the results are negative and 4 are zero. Sample volume, in grams, is provided for 605 (6%) of the results. MDCs are not provided for TIMS analyses.

5.3.6 Post-1991 *In Vitro* Data for ^{241}Am

The post-1991 *in vitro* data table contains 2,657 records for ^{241}Am analyses. These are summarized in Table 5-34.

Table 5-34. Summary for ^{241}Am records in the post-1991 *in vitro* data.

Collection requirement	Collection protocol (no. records)	Collection date range	Analysis types (no. records)
Prompt action (228 records)	Simulated 24 hour (77)	3/30/1994–11/29/2008	RAS (60), PHW (17)
	Spot fecal (151)	8/8/1989–3/10/2005	RAS (93), PHW (58)
Special (436 records)	Simulated 24 hour (417)	5/26/1991–12/14/2008	RAS (417)
	Spot fecal (18)	12/29/1973–8/3/2005	RAS (18)
	True 24 hour (1)	8/20/1997	RAS (1)
Routine (1,993 records)	Simulated 24 hour (1,993)	5/9/1991–1/19/2009	RAS (1,993)

1,921 (96%) of the 1,993 routine urinalysis records for ^{241}Am have a non-null result in the database. Results are reported for all three result types, as shown in Table 5-35. For the results given in terms of excretion, 18 are reported in units of pCi per sample. The rest are in pCi per 24 hours.

Table 5-35. Non-null results for routine ^{241}Am analyses in the post-1991 *in vitro* data.

Result type	No. non-null records	No. records less than zero	No. records = zero
Per aliquot	882	521	2
Per sample	290	75	1
Excretion basis	749	346	3

Sample volumes are reported for 93% of the non-null per aliquot results, for 100% of the per sample results, and for 88% of the excretion results. Sample volumes are given primarily in grams, with some values given in mL. All of the per aliquot and per sample results have MDCs. MDCs are reported for 88% of the excretion results, in units of either pCi per aliquot (53%) or pCi per sample (47%). None of the MDC values are zero or negative.

6.0 VALIDATION OF TUPO DATA AGAINST LABORATORY NOTEBOOKS

The archived TUPO data had been entered from laboratory notebooks, which were the formal record for personnel bioassay results before about 1980. Laboratory notebooks were issued to individuals and were often used as journals rather than being dedicated to a specific process or procedure. Consequently, the original bioassay data are distributed across over 100 notebooks, where an individual notebook might contain several thousand records or only a few depending on how it was used by the assignee.

Many of the VAX data files included fields that identified the laboratory notebooks from which the bioassay data originated. A cursory comparison between the VAX data files and several of the notebooks suggested a rather high rate of disagreement (~20%). This prompted an effort to validate a larger fraction of the TUPO data to gain a better understanding of its reliability.

From the VAX data files, 120 laboratory notebooks were identified as containing TUPO data. Ninety of these were retrieved, which represented nearly 90% of the TUPO records.

In reviewing the data, priority was placed on validating positive results and high values in particular. However, the review also included a considerable fraction of results for which no activity (i.e., 0.00) was reported. Errors were found in the reporting of sample results, sample dates, and notebook and page references. Ninety-six records were added to the database that had been omitted from the VAX files.

Eleven percent (16,692 of 147,690) of the TUPO records were validated. Data comments were entered for 4,882 records, and 1,330 records were corrected in some way. The majority (83%) of the corrections were changes to notebook or page references. Beyond corrections, data comments were used to document information such as unresolved questions about a record or errors that were apparent but could not be corrected, such as an incorrect notebook reference where the correct notebook number could not be determined. Table 6-1 summarizes the validation results and the types of errors found.

Table 6-1. Data validation summary for TUPO records.

Total records	Validated records	Records added	Corrected fields				Data comments
			Sample results	Sample date	Notebook reference	Page reference	
147,690	16,692	102	209	12	578	531	4,882

Data validation issues specific to tritium, uranium, and polonium records are discussed in Sections 6.1 through 6.3. Section 6.4 describes the review of the laboratory notebooks for bioassay data for other radionuclides. This was performed concurrently with the validation of the TUPO data.

6.1 VALIDATION OF TRITIUM RECORDS

From the VAX files, 27 notebooks were identified as containing tritium bioassay data. Fifteen of these were obtained, which represented 35,267 tritium records or 70% of the total. A sixteenth notebook (number S4819) was retrieved, but it did not contain tritium bioassay information. There was a significant period (1958 to 1968) for which notebooks were either not found or could not be identified. Therefore, none of the tritium bioassay data from this period have been validated. The fifteen notebooks that were used in the validation of tritium records from the VAX files are listed in Table 6-2.

Approximately 10% (5,150 out of 50,141) of the tritium records were validated. These totals include 34 records added to the database that were present in the notebooks but not in the VAX files. Data

Table 6-2. Laboratory notebooks used to validate *in vitro* analytical results for tritium.

Notebook number	Units used	Start date	End date	No of records	Percent validated	Comments
3605	μCi/L	06/23/50	09/11/52	1,582	18	Contains earliest tritium data and calibration details on pages 1–8 from which some uncertainty information might be derived.
5481	μCi/L	09/11/52	09/19/54	5,148	6.6	
6813	μCi/L	09/17/54	12/30/57	5,517	6.8	Contains memorandum in front that might help in determining uncertainty from 4/20/50 to 01/66.
15722	μCi/L	01/03/69	01/24/72	3,495	4.6	On page 16 there is a change in reporting. Values reported as "<1" are now reported well below this value. The VAX files report results 1% to 10% higher than reported in this notebook.
17487	μCi/L	01/25/72	11/08/74	2,283	7.4	Page 1 has some statements on accuracy and precision.
18519	μCi/L	11/07/74	08/27/76	1,772	19.5	
20096	μCi/L	08/25/76	09/27/78	1,812	6.6	
21536	μCi/L	09/26/78	04/02/80	2,069	7	
22311	μCi/L	03/28/80	07/14/81	1,778	8.3	
25285	μCi/L	10/29/82	03/05/85	1,353	10	
24959	μCi/L	02/21/84	05/15/84	1,774	5.1	
24906	μCi/L	05/28/85	12/10/87	2,496	8	
R6066	μCi/L	12/11/86	01/28/88	1,933	8	
R6074	μCi/L	01/19/88	07/07/89	2,232	6.2	
R7709	μCi/L	12/21/90	06/24/91	23	100	

comments were made for 3,564 records, and corrections were applied to 110 records. Validation of the tritium data is summarized in Table 6-3.

Table 6-3. Tritium data validation summary.

Total records	Validated records	Records added	Corrected fields				Data comments
			Sample results	Sample date	Notebook reference	Page reference	
50,141	5,150	34	45	2	12	51	3,564

In comparing the data in the notebooks with that in the VAX files, it was evident that the bioassay results from notebook 15722 had been adjusted in some manner. In general, the values from the VAX files were 1% to 10% higher than those documented in the notebook. There were approximately 3,460 affected records. This discrepancy was indicated in the data comment field.

6.2 VALIDATION OF URANIUM RECORDS

From the VAX files, 83 notebooks were identified as possibly containing *in vitro* bioassay data for uranium. Sixty-four of these were retrieved, and 58 were determined to contain uranium bioassay data. The 58 notebooks contained 85,857 uranium records, representing 93% of the total. The notebooks are listed in Table 6-4.

Table 6-4. Laboratory notebooks used to validate *in vitro* analytical results for uranium.^a

Notebook	Record type and units	Start date	End date	No. of records	Percent validated	Comments
11246	U-235 (dpm/L)	08/16/63	04/15/65	4,353	2.8	
13764	U-235 (dpm/L)	04/23/65	04/17/67	4,571	3.4	
13833	U-235 (dpm/L)	04/17/67	11/18/68	3,579	3.5	

Notebook	Record type and units	Start date	End date	No. of records	Percent validated	Comments
15721	U-235 (dpm/L and counts per 50 minutes)	11/22/68	09/08/70	3,683	3.5	Beginning with about p. 50, the results are reported in counts per 50 minutes. These were adjusted for efficiency, sample size, and count time to get dpm/L, the reporting units for the computer data files.
16283	U-235 (µg/L)	09/14/70	05/05/72	1,491		These results had been adjusted in some undetermined way between the notebook values and those in the VAX files. Since it was not clear how the values had been adjusted, validation of the data in this notebook was impracticable.
17494	U-235 (dpm/L)	05/19/72	10/19/73	1,706	6	These results were adjusted for efficiency, sample size, and count time to get dpm/L, the reporting units for the computer data files. (Size = 75 ml, Count time = 50 min, Recovery = 0.7, and Efficiency = 0.4)
18511	U-235 (dpm/L)	11/02/73	07/04/75	1,907	2	These results were adjusted for efficiency, sample size, and count time to get dpm/L, the reporting units for the computer data files. (Size = 75 ml, Count time = 50 min, Recovery = varies, and Efficiency = 0.4)
6654	U-235			678		Contains air samples, blood samples for toxic exposures, and U-235.
8949	U-235 (dpm/L)	07/16/62	08/16/63	1,714	4.2	
9463	U-235 (dpm/L)	09/21/58	07/16/62	2,341	3.8	
S2179	U-235 (dpm/L until 1/77, then pCi/L)	07/18/75	09/13/77	2,128	4.9	
S3436	U-235 (pCi/L) and U-238 (µg/L)	10/30/81	02/05/82	1,418	6.7	
19947	U-235 (pCi/L) and U-238 (µg/L)	09/12/77	03/12/79	3,443	7.5	
21533	U-235 (pCi/L) and U-238 (µg/L)	09/21/79	01/25/80	1,891	10	
22314	U-235 (pCi/L) and U-238 (µg/L)	06/02/80	09/19/80	1,555	5.6	
22316	U-235 (pCi/L) and U-238 (µg/L)	10/06/81	01/23/81	1,560	3	
22318	U-235 (pCi/L) and U-238 (µg/L)	02/11/80	05/16/80	1,220	8.8	
22319	U-235 (pCi/L) and U-238 (µg/L)	01/25/80	02/16/81	19	100	
23145	U-235 (pCi/L) and U-238 (µg/L)	06/12/81	10/16/81	1,577	20	
23148	U-235 (pCi/L) and U-238 (µg/L)	02/06/81	06/12/81	1,812	4.7	
24082	U-235 (pCi/L) and U-238 (µg/L)	06/25/82	12/24/82	708	14	
24907	U-235 (pCi/L) and U-238 (µg/L)	11/13/84	06/24/85	812	20	
24912	U-235 (pCi/L) and U-238 (µg/L)	03/12/84	10/29/84	948	13	
24951	U-235 (pCi/L) and U-238 (µg/L)	01/07/83	07/08/83	822	31	
24955	U-235 (pCi/L) and U-238 (µg/L)	07/14/83	03/05/84	966	8.2	
A1185	U-235 (pCi/L) and U-238 (µg/L)	06/24/85	02/18/86	797	38	
R6063	U-235 (pCi/L) and U-238 (µg/L)	06/08/88	06/22/90	1,010	26	
R6070	U-235 (pCi/L) and U-238 (µg/L)	09/29/86	06/08/87	786	17	

Notebook	Record type and units	Start date	End date	No. of records	Percent validated	Comments
R6574	U-235 (pCi/L) and U 238 (µg/L)	06/22/87	04/18/88	772	21.5	
S2517	U-235 (pCi/L) and U 238 (µg/L)	03/23/79	09/07/79	2,820	2.4	
S4412	U-235 (pCi/L) and U 238 (µg/L)	01/08/82	06/11/82	1,396	21	
S9787	U-235 (pCi/L) and U 238 (µg/L)	03/03/86	09/15/86	656	38	
17493	U-238 (µg/L)	02/01/77	10/31/77	20	100	
18510	U-238 (µg/L)	11/09/73	10/19/76	1,682		These results had been adjusted in some undetermined way between the notebook values and those in the VAX files. Since it was not clear how the values had been adjusted, validation of the data in this notebook was impracticable.
3064	U-238 (µg/L)	11/18/49	04/22/50	248	68	
3351	U-238 (µg/L)	03/24/50	09/09/50	400	54.5	
3683	U-238 (µg/L)	07/29/50	09/05/59	714	16	
3802	U-238 (µg/L)	08/16/59	01/06/51	574	56	
3832	U-238 (µg/L)	09/25/50	02/06/52	118	23	
4992	U-238 (µg/L)	01/14/52	02/16/53	953	16	
5843	U-238 (µg/L)	01/19/53	06/05/53	911	13	
6438	U-238 (µg/L)	10/13/53	04/07/54	1,101	7	
6744	U-238 (µg/L)	04/12/53	08/19/54	1,181	8.4	
6982	U-238 (µg/L)	08/20/54	01/10/55	1,136	5.9	
R108	U-238 (µg/L)	01/07/55	02/17/56	2,473	4	
S1068	U-238 (µg/L)	08/20/64	06/17/66	1,926	7	
S1199	U-238 (µg/L)	02/02/76	05/30/78	2,276	5.4	
S1214	U-238 (µg/L)			1,162		
S1228	U-238 (µg/L)	07/15/66	11/13/67	1,735	4.6	
S1474	U-238 (µg/L)	03/14/69	08//28/70	1,273	4.6	
S1653	U-238 (µg/L)	09/16/70	12/02/70	1,084	6.7	
S1790	U-238 (µg/L)	12/03/71	10/12/73	1,673		These results had been adjusted in some undetermined way between the notebook values and those in the VAX files. Since it was not clear how the values had been adjusted, validation of the data in this notebook was impracticable.
S2521	U-238 (µg/L)	06/19/78	02/22/80	569	44	
S447	U-238 (µg/L)	03/15/57	08/14/58	1,525	3.6	
S615	U-238 (µg/L)	08/22/58	02/05/60	1,675	11	
S727	U-238 (µg/L)	02/18/60	03/17/61	1,062	3	
S792	U-238 (µg/L)	04/07/61	08/03/62	1,392	8	
S901	U-238 (µg/L)	08/15/62	08/07/64	1,855	4	

a. LANL refers to enriched uranium as "U-235" and other forms, including normal uranium, as "U-238." The use of "U-235" and "U-238" in this context does not imply isotope-specific analyses.

There were 17 ²³⁵U and 9 ²³⁸U records in the notebooks that did not appear in the VAX files. These were added to the database. In addition, six ²³⁸U records were found in the file "sPOL4765.dat" that had been cataloged as containing only polonium data. The fact that these six records were uranium rather than polonium assay results was verified through examination of the laboratory notebooks. These six ²³⁸U records were also added to the database.

Corrections were made to 122 ²³⁵U records and comments were made for 76. For ²³⁸U, corrections were applied to 78 records and comments were made for 803. Validation of the uranium data is summarized in Table 6-5.

Table 6-5. Uranium data validation summary.

Nuclide	Records	Validated records	Records added	Corrected fields				Data comments
				Sample results	Sample date	Notebook reference	Page reference	
U-235	43,668	4,436	17	32	0	1	79	76
U-238	48,169	4,841	15	60	10	2	6	803
Total	91,837	9,277	32	92	10	3	85	879

6.3 VALIDATION OF POLONIUM RECORDS

The VAX files identified 13 notebooks with polonium bioassay data. Nine of these were retrieved of which three were determined to contain bioassay data for polonium. The three notebooks contained 5,682 polonium records, which represented 99% of the total. The retrieved notebooks are listed in Table 6-6.

Table 6-6. Laboratory notebooks used to validate *in vitro* analytical results for polonium.

Notebook	Start date	End date	Units codes used	Number of records	Percent validated
1794A	03/11/47	01/09/53	CM: cpm/24 hr DM: dpm/L	2,957	33
3174	01/09/53	03/24/65	DM: dpm/L NCl: nCi/g fecal	1,726	66
H5100	12/31/51	12/15/53	CM: cpm/24 hr DM: dpm/L	986	10

Approximately 39% of the polonium records (2,245 of 5,712) were validated. There were 36 records that were added to the database that were present in the notebooks but not in the VAX files. Corrections and comments were made for 1,020 and 439 records, respectively. All but 72 of the 1,020 corrections were for notebook or page reference issues. Validation of the polonium data is summarized in Table 6-7.

Table 6-7. Polonium data validation summary.

Total records	Validated records	Records added	Corrected fields				Data comments
			Sample results	Sample date	Notebook reference	Page reference	
5,712	2,245	36	72	0	553	395	439

6.4 REVIEW OF LABORATORY NOTEBOOKS FOR *IN VITRO* MONITORING DATA FOR OTHER RADIONUCLIDES

In the process of validating the TUPo data against laboratory notebooks, around 100 to 200 bioassay records were identified where LANL had performed special analyses for other radionuclides, such as ⁹⁰Sr. These records were not included in the bioassay database because they were not part of LANL's formal bioassay program and there was too much uncertainty about their origin or reliability.

Review of the laboratory notebooks did not identify any bioassay data for LANL employees who worked with radioactive lanthanum (RaLa). Workers who handled RaLa or its waste streams had the potential for intakes of ⁹⁰Sr, that was present as a contaminant. A small number of gross beta results for ⁹⁰Sr were identified, but it did not appear there was ever a formal bioassay program for ⁹⁰Sr.

Review of the notebooks also identified isolated instances of *in vitro* bioassay for mixed fission products, but as in the ⁹⁰Sr cases these data were not associated with a formal bioassay program. There are cases where *in vivo* counting was performed for mixed fission products.

7.0 ATTRIBUTIONS AND ANNOTATIONS

All information requiring identification was addressed via references integrated into the reference section of this document.

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ATTACHMENT A URANIUM NEUTRON ACTIVATION ANALYSIS

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Introduction

This attachment presents information about LANL's UNAA urinalysis program. UNAA was one of the uranium analysis types encountered during the development of the LANL bioassay database. Use of UNAA was implemented in the late 1970s and ceased in 1990. (There is a single result in the database from 1962 that shows UNAA as the analysis type, but this is thought to be a data entry error.) UNAA involved irradiating a urine specimen in a thermal neutron beam and subsequently counting it for delayed neutrons from fission product precursors.

UNAA Bioassay Program

As of 1978 UNAA was an element of LANL's routine bioassay program. LANL's experience with the UNAA method had shown that samples giving a neutron count greater than the detection limit of the neutron counting system (at 99% confidence) also showed positive uranium concentrations when analyzed by wet chemistry. The method therefore provided a way to screen a large number of samples, with subsequent wet chemistry analyses performed as needed to confirm high values or determine the $^{235}\text{U}/^{238}\text{U}$ ratio (Ide et al. 1979).

Personnel were entered into the UNAA bioassay program based on information from their Health Physics Checklist (HPC). The HPC documented if an individual was going to be working with ^{235}U (enriched uranium), ^{238}U (depleted or normal uranium), or both.

As of 1978 LANL's action levels (which they called "concern levels,") for uranium in urine were 10 $\mu\text{g}/\text{L}$ for depleted and natural uranium, and 4.5 pCi/L for enriched uranium (Ide et al. 1979).

UNAA Calibration and Detection Limits

The counting method was calibrated by sending urine samples spiked with known quantities of uranium through the analysis process. Urine standards were prepared using depleted uranium (0.18 atom percent ^{235}U), normal uranium (0.72 atom percent ^{235}U), and enriched uranium (93.16 atom percent ^{235}U). Standards were prepared over a range of uranium concentrations for each enrichment: 0 to 2,500 $\mu\text{g}/\text{L}$ for depleted uranium, 0 to 140 $\mu\text{g}/\text{L}$ for normal uranium, and 0 to 1,000 pCi/L for enriched uranium (Ide et al. 1979).

LANL determined that the irradiation of a 25 cm^3 urine specimen containing 4.16 $\mu\text{g}/\text{L}$ of depleted uranium (0.18 atom percent ^{235}U) would give a gross neutron count of 100 counts, which was the detection limit of the counting system at 99% confidence. The 100-count detection limit was determined from counting statistics, using a background established from a series of urine control samples (Ide et al. 1979). The MDCs that corresponded to the 100-count detection limit for the UNAA method are given in Table A-1 for depleted, natural, and enriched uranium.

Table A-1. MDCs for UNAA urinalysis at LANL (Ide et al. 1979).

Uranium enrichment	MDC
Depleted uranium (0.18 atom percent ^{235}U)	4.16 $\mu\text{g}/\text{L}$
Normal uranium (0.72 atom percent ^{235}U)	1.04 $\mu\text{g}/\text{L}$
Enriched uranium (93.16 atom percent ^{235}U)	1.5 pCi/L

UNAA Analyses and Reporting Units

Samples were collected from personnel by the LANL industrial hygiene group (H-5 at the time). Neutron irradiation and counting took place at the Omega West Reactor facility. Quality control samples and blanks were analyzed concurrently with the routine unknown samples to allow corrections to be made for changes in background or reactor power (Ide et al. 1979).

ATTACHMENT A
URANIUM NEUTRON ACTIVATION ANALYSIS

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The UNAA process involved irradiating 25 cm³ urine specimens in a thermal neutron beam for 60 seconds and then counting them for delayed neutrons for a 55-second interval following a short decay period (Ide et al. 1979). The delayed neutron count was equated to uranium concentration using an assumed ²³⁵U fraction (i.e., enrichment) that reflected an individual's exposure environment. (Delayed neutron assay methods cannot differentiate between fissile species or varying uranium enrichments, so one must have prior knowledge or use assumptions to quantify results.)

Results for depleted or normal uranium were denoted as "²³⁸U" and reported in micrograms per liter. Results for enriched uranium were denoted as "²³⁵U" and reported in picocuries per liter.