

**Revised Methodology for Determination of Inventories in SDF
Vaults 1 and 4 through 9/30/2011**


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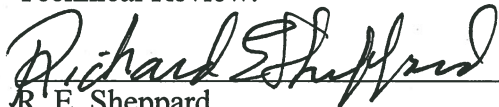
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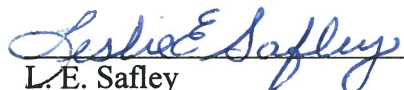
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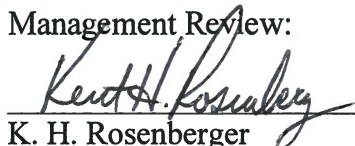
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TABLE OF CONTENTS

LIST OF FIGURES	4
LIST OF TABLES	4
ACRONYMS	5
EXECUTIVE SUMMARY	6
1.0 BACKGROUND	6
2.0 CURRENT SDF INVENTORY ESTIMATE	8
3.0 INVENTORY UPDATE	8
3.1 Sample Determination	8
3.2 Special Methods and Assumptions.....	9
3.3 Corresponding Sample and Transfer Data.....	22
3.4 Decay Calculations	25
3.5 Updated Inventories.....	25
4.0 CONCLUSIONS.....	26
5.0 REFERENCES	27
APPENDIX A	30
APPENDIX B	39

LIST OF FIGURES

Figure 1.0-1: Cs-137 Activity Concentrations.....	7
Figure 3.1-1: Data Point Determination Flow Chart	9
Figure 3.3-1: Tank 50 Volumes and Sample Dates	22

LIST OF TABLES

Table 3.3-1: Transfer Periods and Sample Dates for Vault 1	23
Table 3.3-2: Transfer Periods and Sample Dates for Vault 4	24
Table 3.5-1: Updated Vault 1 Inventory	25
Table 3.5-2: Updated Vault 4 Inventory	26
Table A.0-1: Radionuclide Activity Concentrations by Sample Date	30
Table B.0-1: Cs-135 and Cs-137 Reported Concentrations in pCi/mL	39

ACRONYMS

ARP	Actinide Removal Process
DDA	Dissolution Deliquification and Adjustment
DSS	Decontaminated Salt Solution
FY	Fiscal Year
MCU	Modular Caustic Side Solvent Extraction
ISDP	Interim Salt Disposition Processing
SDF	Saltstone Disposal Facility
SDU	Saltstone Disposal Unit
SPF	Saltstone Production Facility
SRS	Savannah River Site
WCS	Waste Characterization System

EXECUTIVE SUMMARY

The Saltstone Disposal Facility (SDF) at the Savannah River Site (SRS) is the site of disposal of low-level liquid waste that has been mixed with cement, fly ash, and blast furnace slag to create a stable waste form known as saltstone. This saltstone is stored in Saltstone Disposal Units (SDUs) engineered to contain the radionuclides within the saltstone matrix. The SDUs that currently contain saltstone include Vault 1 and Vault 4.

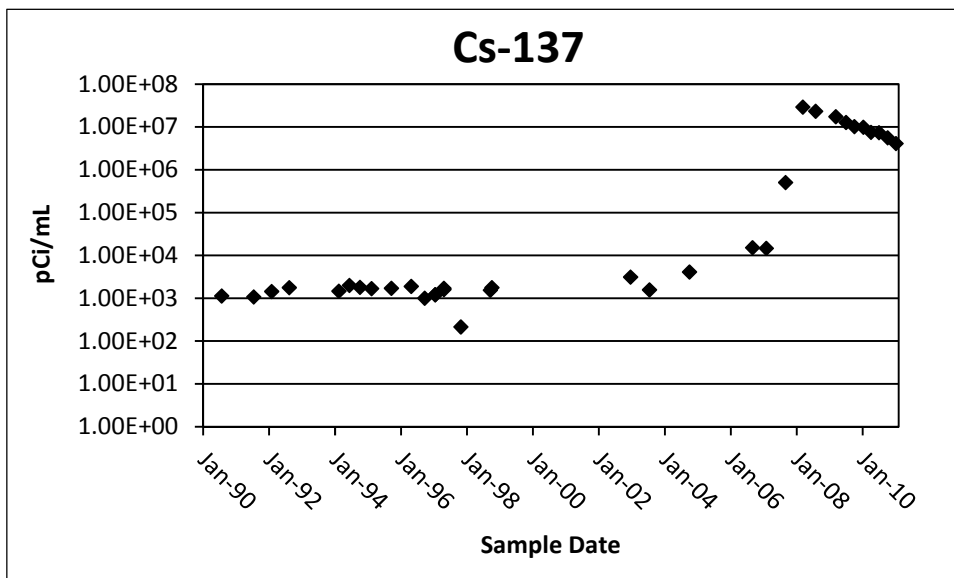
Historically, a running radiological inventory has been calculated annually based on samples from the feed tank (Tank 50), material balances, assumed influent stream concentrations, and the site Waste Characterization System (WCS). Due to the high level of conservatism in some of these sources compounded over years of facility operation, the current reported radiological inventory is not truly indicative of the SDF inventory. This effort uses a systematic approach to remove unnecessary conservatism and give a more accurate picture of the SDF radiological inventory (as of 9/30/2011) as a basis for future inventory calculation.

1.0 BACKGROUND

Disposal of salt solution at SRS began in June of 1990 with very low-activity liquid waste and has continued until present day. Salt solution is processed via the Saltstone Production Facility (SPF) into liquid grout, which is then deposited in SDUs within the SDF where it hardens into saltstone. The SDUs currently containing saltstone are known as Vault 1 and Vault 4. Vault 1 contains the earliest and lowest activity waste, while Vault 4 contains some of the slightly higher activity decontaminated salt solution (DSS) from Interim Salt Disposition Processing (ISDP) including Deliquification, Dissolution and Adjustment (DDA) and Actinide Removal Process/Modular Caustic Side Solvent Extraction Unit (ARP/MCU).

Saltstone made from DSS processed using only DDA has the highest activity, as later ARP/MCU DSS experiences DDA processing as well as removal of targeted radionuclides such as cesium, strontium, and actinides. From the beginning of DDA processing, Tank 50 has never been completely emptied, resulting in continued dilution of the DDA material in Tank 50. This can be seen clearly in the Cs-137 activity concentrations over time, shown in Figure 1.0-1. Similar charts for all radionuclides can be found in Appendix A.

Figure 1.0-1: Cs-137 Activity Concentrations



This assessment of inventory relies, whenever possible, on sample detection information from the feed material, salt solution in Tank 50. Due to the limitations of analytical methods and the large number of reported radionuclides, many of the values reported in inventory calculations are based on detection limits, special methods or assumed values. Detection limits provide a bounding value that gives the limit of the detection method. This value is greater than the true activity concentration, sometimes by orders of magnitude. Thus, this assessment uses special methods and assumed activity concentrations to reduce this conservatism where appropriate. As discussed in Section 3.2, special methods and assumed values are based on relationships between radionuclides such as equilibrium relationships, decay chains, and/or process knowledge.

2.0 CURRENT SDF INVENTORY ESTIMATE

The previous SDF inventory had been calculated annually based on contributions to the inventory of the current year added to an inventory from the most recent annual report decayed one year. Quarterly reports are issued with information about Tank 50 sample results, salt solution transferred to SPF, grout disposed, remaining volume, and total curie inventory information. These reports rely, in part, on activity concentration data taken from WCS reporting and streams with assumed concentrations that are introduced into Tank 50. Some of these concentrations are assumed to be at the WAC limit for conservatism, which is detrimental to establishing an accurate radiological inventory of material disposed. Inventory in these annual reports is also decayed as if the entire inventory is added at the end of the current fiscal year (FY), which is conservative for radionuclides with short half-lives but can underestimate contributions due to ingrowth of decay products.

The most recent annual summary (X-CLC-Z-00034) estimated the total inventories in Vault 1 and Vault 4 (as of 9/30/2010) to be 191 curies and 488 kilocuries respectively. This document recalculates both inventories using Tank 50 activity concentration values and SPF transfer volumes, and then decays the resulting inventory from the original quarter of disposal.

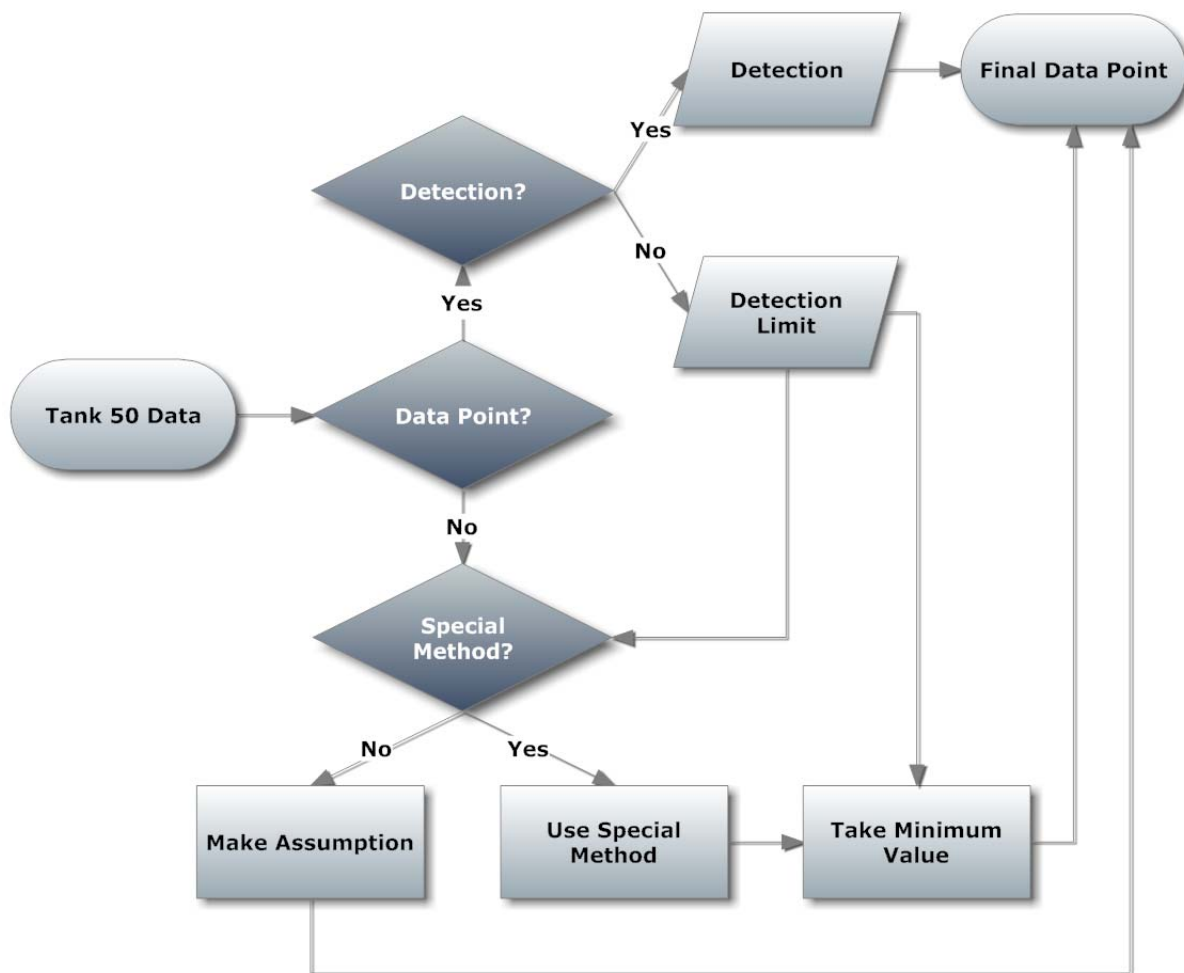
3.0 INVENTORY UPDATE

In order to correct some of the conservatism in the SDF radiological inventory, historical Tank 50 sample information was compiled from the beginning of SPF processing, with the earliest sample being from July of 1990. Missing information was calculated based on documented special methods or assumed based on process information. Transfer volume data was also compiled from the beginning of processing using the volumes reported transferred from Tank 50 to the SPF. The inventory resulting from the combination of these transfers and corresponding representative samples were grouped by calendar quarter of disposal and decayed to 9/30/2011.

3.1 Sample Determination

Due to the large number of radionuclides of widely varying activities, detection is not always possible. Inventory for each radionuclide is based whenever possible on detections, with the choice of activity concentration used following the flow chart in Figure 3.1-1.

Figure 3.1-1: Data Point Determination Flow Chart



Analysis for each radionuclide began with analysis of the Tank 50 sample data. Data points were identified as detections or detection limits. Detections were always used as the final activity concentration. If the data point was a detection limit with no special method, the detection value itself was used. If there was a special method associated with a detection limit, then the two were compared and the minimum value was taken. If there was no detection or detection limit, the final data point was taken from either a special method or an assumption. These special methods and assumptions are detailed by radionuclide in Section 3.2.

3.2 Special Methods and Assumptions

In determining which activity concentrations to use, effort was made either to develop assumptions based on known sample data and tank farm inventories or to use existing methods of estimation based on decay or ratio calculations. Detections, as the highest caliber data, were always used with missing data populated using assumptions and/or special methods, as described in Figure 3.1-1. In order to remove unnecessary conservatism in inventory reporting, where both special methods and detection limits existed, the minimum of the two was used. For the purposes of these special methods and assumptions, unless otherwise noted, DDA processing began in January 2006 (this provides a conservative assumption as DDA activity concentrations

tend to be higher than pre-DDA activity concentrations). Note that some assumptions made (where noted) are applicable only to earlier disposal (before DDA) and do not necessarily predict future activity concentrations.

H-3

H-3 needed no special method. The detection limit was used when there was no detection.

C-14

C-14 needed no special method. The detection limit was used when there was no detection.

Na-22

For Na-22, the minimum of the detection limit and a decay calculation based on a value of $5.4\text{E}+03$ pCi/mL was used. Na-22 has a short half-life (approximately 2.6 years), and thus decay was factored into the calculation. A total inventory estimate from 2005 (CBU-PIT-2005-00263) divided over the total expected salt solution volume of $9.9\text{E}+07$ gallons (SRR-LWP-2009-00001) resulted in an assumed value that was unreasonably high based on recent data. This estimate was reduced by a factor of four to produce a more reasonable but still conservative estimate of historical Na-22 sample information.

$$\text{Na-22 Activity Concentration (pCi/mL)} = 5.4 \times 10^{+03} e^{-0.266(Y-2005)}$$

Al-26

For Al-26, when no detection or detection limit was reported, was assumed to have an activity concentration of $3.4\text{E}+01$ pCi/mL using a total expected salt solution volume of $9.9\text{E}+07$ gallons (SRR-LWP-2009-00001) and a soluble inventory of 12.9 curies. [CBU-PIT-2005-00263]

Cl-36

Based on possible sources of Cl-36, its concentration in the waste should be minimal, but historically no analysis for Cl-36 has been performed. Recently activity concentrations for Cl-36 and K-40 have been based on the minimum concentration, which can be confirmed. The detection capabilities are based on the removal of Sr-90. Therefore, the expected minimum detection limit for Cl-36 was based on the concentration of Sr-90. [SRNS-J2100-2008-00004] If additional information is made available on the total Cl-36 inventory or Cl-36 concentrations in the future, this method may be revised. The minimum of the special method and detection limit was used.

$$\text{Cl-36 Activity Concentration (pCi/mL)} = 1.0 \times 10^{-05} * \text{Sr-90 Activity Concentration}$$

K-40

Based on possible sources of K-40, its concentration in the waste should be minimal, but historically little or no analysis for K-40 has been performed, and therefore the special method activity concentration for K-40 has been based on the minimum concentration that could be detected. The detection capabilities were based on the removal of Sr-90. Therefore, the expected minimum detection limit for K-40 was based on the concentration of Sr-90. The minimum of the detection limit and special method was used. Recently, reported detection limits have been an order of magnitude below previously assumed detection limits (four orders of magnitude less than Sr-90). [SRNS-J2100-2008-00004] Therefore, the following relationship is used.

$$\text{K-40 Activity Concentration (pCi/mL)} = 1.0 \times 10^{-05} * \text{Sr-90 Activity Concentration}$$

Co-60

Co-60 needed no special method. The detection limit was used when there was no detection.

Ni-59

The special method activity concentration for Ni-59 was derived from the Ni-63 activity concentration. When both a detection limit and special method value were available, the minimum of the two was used. [SRNL-L3100-2009-00189]

$$\text{Ni-59 Activity Concentration (pCi/mL)} = 2.0 \times 10^{-02} * \text{Ni-63 Activity Concentration}$$

Ni-63

Ni-63 needed no special method. The detection limit was used when there was no detection.

Se-79

Se-79 needed no special method. The detection limit was used when there was no detection.

Sr-90

The special method activity concentration for Sr-90 was derived, when necessary, from the Cs-137 activity concentration. The minimum of the detection limit and special method was used when there was no detection. [X-ESR-G-00004]

$$\text{Sr-90 Activity Concentration (pCi/mL)} = 2.6 \times 10^{-04} * \text{Cs-137 Activity Concentration}$$

Y-90

Y-90 is a daughter of and in equilibrium with Sr-90, so the activity concentration used was the same as that of Sr-90.

$$\text{Y-90 Activity Concentration} = \text{Sr-90 Activity Concentration}$$

Zr-93

Zr-93 is a parent of and in equilibrium with Nb-93m (in about 120 years), so the activity concentration was assumed the same as Nb-93m.

$$\text{Zr-93 Activity Concentration} = \text{Nb-93m Activity Concentration}$$

Nb-93m

Nb-93m activity concentrations were detections or detection limits with missing data populated by an assumed activity concentration of 1.0E+02 pCi/mL. This assumption is based on the available sample data. Concentrations of other constituents significantly increased since 2007. Since the recent concentrations were slightly greater than 1E+02 and the expectation is that concentrations prior to 2007 would be significantly less than current concentrations, the assumed concentration is believed to be conservative. Note that this assumption is not necessarily applicable to future disposal actions.

Nb-94

Nb-94 needed no special method. The detection or detection limit value was used.

Tc-99

Tc-99 needed no special method. The detection or detection limit value was used.

Ru-106

Ru-106 needed no special method. The detection or detection limit value was used.

Rh-106

Rh-106 is a daughter of and in equilibrium with Ru-106, so the activity concentration used was the same as that of Ru-106.

$$\text{Rh-106 Activity Concentration} = \text{Ru-106 Activity Concentration}$$

Pd-107

Pd-107 is not included in the quarterly characterization of salt solution in Tank 50, so its concentration is based on the generated quantity of Pd-107 computed in CBU-PIT-2005-00178 (411 Curies), an assumed solubility of 0.1 %, and a total expected salt solution volume of 9.9E+07 gallons. [SRR-LWP-2009-00001] This yields an assumed concentration of 1.09 pCi/mL.

Sn-126

Sb-126 needed no special method. The detection or detection limit value was used.

Sb-125

Sb-125 needed no special method. The detection or detection limit value was used.

Sb-126

The special method activity concentration for Sb-126 is derived from the Sb-126m activity concentration (14 % of Sb-126m decays produce Sb-126). The minimum of the detection limit and special method was used. [CBU-PIT-2005-00034]

$$\text{Sb-126 Activity Concentration (pCi/mL)} = 1.4 \times 10^{-01} * \text{Sb-126m Activity Concentration}$$

Sb-126m

Sb-126m is a daughter of and in equilibrium with Sn-126, so the activity concentration used was equal to that of Sn-126. [CBU-PIT-2005-00034]

$$\text{Sb-126m Activity Concentration} = \text{Sn-126 Activity Concentration}$$

Te-125m

The special method activity concentration for Te-125m is derived from the Sb-125 activity concentration (based on decay of Sb-125). The minimum of the detection limit and special method was used. [CBU-PIT-2005-00034]

$$\text{Te-125m Activity Concentration (pCi/mL)} = 2.4 \times 10^{-01} * \text{Sb-125 Activity Concentration}$$

I-129

I-129 needed no special method. The detection or detection limit value was used.

Cs-134

Cs-134 was assumed to have an activity concentration of 10 pCi/mL (corrected for decay) when no detection or detection limit values were available. This assumption was based on the available sample data and was corrected for decay, due to the short half-life (2.06 years) of Cs-134. The one actual detection of Cs-134 recorded was less than this assumption. The pre-DDA concentrations should be lower than those after DDA began. Based on the sample date (7/3/2003) of the one actual detection (6.07 pCi/mL), the decay calculation uses the following expression where *Y* represents the year:

$$\text{Cs-134 Activity Concentration (pCi/mL)} = 1.0 \times 10^{+01} e^{-0.335(Y-2004)}$$

Cs-135

The special method activity concentration for Cs-135 was derived from the Cs-137 activity concentration based on regression of available sample data. In this case, the calculated special method value was compared to the detection limit and the minimum value was used. A basic regression analysis was performed, grouping the data before and after the large activity concentration change in 2008 and correlating Cs-137 with available Cs-135 data, which was used to predict additional Cs-135 activity concentrations. Additional details of these calculations can be found in Appendix B.

Prior to 03/11/2008:

$$\text{Cs-135 Activity Concentration (pCi/mL)} = 1.0 \times 10^{-04} * \text{Cs-137 Activity Concentration} + 13.6$$

03/11/2008 and forward:

$$\text{Cs-135 Activity Concentration (pCi/mL)} = 6.11 \times 10^{-06} * \text{Cs-137 Activity Concentration} + 7.24$$

Cs-137

Cs-137 needed no special method. All samples had detection values.

Ba-137m

Ba-137m is a daughter of and in equilibrium with Cs-137, so the activity concentration used was based on that of Cs-137. The equilibrium relationship was used.

$$\text{Ba-137m Activity Concentration (pCi/mL)} = 9.46 \times 10^{-01} * \text{Cs-137 Activity Concentration}$$

Ce-144

The special method activity concentration for Ce-144 was based on a decay calculation. [SRNL-L3100-2009-00189] The minimum of the detection limit and special method was used. As Tank 50 still contains a significant amount of DDA material, it would be conservative to assume the “during DDA” concentrations until after most of that material has been removed from Tank 50, which has not yet occurred. The following equations represent these decay calculations where Y is the year.

During DDA:

$$\text{Ce-144 Activity Concentration (pCi/mL)} = 4.4 \times 10^{+01} e^{-0.888(Y-2009)}$$

End of DDA and forward:

$$\text{Ce-144 Activity Concentration (pCi/mL)} = 1.7 e^{-0.888(Y-2009)}$$

Pr-144

Pr-144 is a daughter of and in equilibrium with Ce-144, so the activity concentration used was the same as that of Ce-144. [SRNL-L3100-2009-00189]

$$\text{Pr-144 Activity Concentration} = \text{Ce-144 Activity Concentration}$$

Pm-147

Pm-147, when no detection or detection limit was available, was assumed to have an activity concentration of 10 pCi/mL (corrected for decay). Note that this assumption was only applied to dates prior to DDA and is not necessarily applicable to future disposal actions. This assumption was based on available sample data and detection limits and the decay was added due to the short (2.62 years) half-life. The decay correction is implemented due to the short half-life of 2.62 years. Based on a detection limit of 8.13 pCi/mL on 10/1/2004. The following expression accounts for decay where Y represents the year:

$$\text{Pm-147 Activity Concentration (pCi/mL)} = 1.0 \times 10^{+01} e^{-0.264(Y-2005)}$$

Sm-151

Sm-151, when no detection or detection limit was available, was assumed to have an activity concentration of 1.0 pCi/mL. This assumption was based on available sample data, detection limits, and sample date. Note that this assumption is not necessarily applicable to future disposal actions.

Eu-152

Eu-152, when no detection or detection limit was available, was assumed to have an activity concentration of 0.1 pCi/mL (corrected for decay). The half-life of Eu-152 is 13.5 years. Note that this assumption is not necessarily applicable to future disposal actions. This assumption was based on available sample data, detection limits, and sample date. A detection of 6.88E-02 pCi/mL occurred on 12/20/2002; thus, the following expression was used:

$$\text{Eu-152 Activity Concentration} = 1.0 \times 10^{-01} e^{-0.0512(Y-2003)}$$

Eu-154

E-154 needed no special method. The detection or detection limit value was used.

Eu-155

Eu-155, when no detection or detection limit was available, was assumed to have an activity concentration of 0.11 pCi/mL (corrected for decay). Note that this assumption is not necessarily applicable to future disposal actions. This assumption was based on available sample data, detection limits, and sample date and was decayed due to the relatively short half-life (4.75 years). A detection of 0.111 pCi/mL occurred on 7/26/2003, so when no detection or detection limit was available the following expression was used:

$$\text{Eu-155 Activity Concentration} = 1.1 \times 10^{-01} e^{-0.146(Y-2004)}$$

Pt-193

Pt-193 was not included in the characterization of salt solution in Tank 50, so its concentration was based on the generated quantity of Pt-193 conservatively computed in LWO-PIT-2006-00039 (1.14E+05 curies), an assumed solubility of 0.1 %, and a total expected salt solution volume of 9.9E+07 gallons (SRR-LWP-2009-00001) based on current and projected SDU volume. This yields an assumed concentration of 3.0E+02 pCi/mL and decay (the half-life of Pt-193 is 50 years) was not considered here.

Ra-226

The special method activity concentration for Ra-226 was based on a decay calculation. [SRNL-L3100-2009-00189] The minimum of the detection limit and special method was used. As Tank 50 still contains a significant amount of DDA material, it would be conservative to assume the “during DDA” concentrations until after most of that material has been removed from Tank 50, which has not yet occurred. The “after DDA” activity concentration calculation should be conservative for the material disposed of before DDA, and was therefore used to predict activity concentrations prior to 2006.

where Y is the year

After DDA:

$$\text{Ra-226 Activity Concentration (pCi/mL)} = 8.3 \times 10^{-22} e^{0.0188Y}$$

During DDA:

$$\text{Ra-226 Activity Concentration (pCi/mL)} = 4.2 \times 10^{-20} e^{0.0188Y}$$

Ra-228

Ra-228 is a daughter of, and was assumed to be in equilibrium with, Th-232, so the activity concentration used was based on that of Th-232. The minimum of the detection limit and equilibrium relationship was used.

$$\text{Ra-228 Activity Concentration} = \text{Th-232 Activity Concentration}$$

Ac-227

Ac-227 is a granddaughter of U-235, so the activity concentration used was based on that of U-235. The minimum of the detection limit and decay relationship was used. [CBU-PIT-2005-00040]

where $t = \text{Year} - 1960$ and $A_i(t)$ is the activity of isotope i at time t

$$A_{Ac-227}(t) = A_{U-235}(t) \left[1 - 1.00066 \left(\frac{1}{2} \right)^{3.0488 \times 10^{-5} t} + 6.6 \times 10^{-4} \left(\frac{1}{2} \right)^{4.5926 \times 10^{-2} t} \right]$$

Th-229

Th-229 is a daughter of U-233, so the activity concentration used was based on that of U-233. The minimum of the detection limit and decay relationship was used. [CBU-PIT-2005-00040]

where $t = \text{Year} - 1960$ and $A_i(t)$ is the activity of isotope i at time t .

$$A_{Th-229}(t) = 1.0481A_{U-233}(t) \left[1 - \left(\frac{1}{2} \right)^{1.307 \times 10^{-4} t} \right]$$

Th-230

Th-230 is a daughter of U-234, so the activity concentration used was based on that of U-234. The minimum of the detection limit and decay relationship was used. [CBU-PIT-2005-00040]

where $t = \text{Year} - 1960$ and $A_i(t)$ is the activity of isotope i at time t

$$A_{Th-230}(t) = 1.442A_{U-234}(t) \left[1 - \left(\frac{1}{2} \right)^{9.1976 \times 10^{-6} t} \right]$$

Th-232

Th-232, when no detection or detection limit was available, was assumed to have an activity concentration of 1.00E-03 pCi/mL. This assumption is based on available sample data and detection limits around the same time. Note that this assumption is not necessarily applicable to future disposal actions.

Pa-231

Pa-231 is a daughter of U-235, so the activity concentration used was based on that of U-235. The minimum of the detection limit and decay relationship was used. [CBU-PIT-2005-00040]

where $t = \text{Year} - 1960$ and $A_i(t)$ is the activity of isotope i at time t .

$$A_{Pa-231}(t) = A_{U-235}(t) \left[1 - \left(\frac{1}{2} \right)^{3.0488 \times 10^{-5} t} \right]$$

U-232

The activity concentration for U-232 was determined first by a detection value, then a detection limit, or, if a detection limit was not reported, the activity concentration was assumed to be 0.11 pCi/mL based on calculations using a total expected salt solution volume of 9.9E+07 gallons (SRR-LWP-2009-00001) and the soluble inventory of 4.11E-02 curies. [CBU-PIT-2005-00263]

U-233

The activity concentration for U-233 was determined first by a detection value, then a detection limit, if a detection limit was not reported, the activity concentration was assumed to be 7.7 pCi/mL based on calculations using a total expected salt solution volume of 9.9E+07 gallons (SRR-LWP-2009-00001) and the soluble inventory of 2.89 curies. [CBU-PIT-2005-00263]

U-234

The activity concentration for U-234 was determined first by a detection value, then a detection limit. If a detection limit was not reported, the activity concentration was assumed to be 12 pCi/mL based on calculations using a total expected salt solution volume of 9.9E+07 gallons (SRR-LWP-2009-00001) and the soluble inventory of 4.43 curies. [CBU-PIT-2005-00263]

U-235

The activity concentration for U-235 was determined first by a detection value, then a detection limit. If a detection limit was not reported, the activity concentration was assumed to be 0.33 pCi/mL based on calculations using a total expected salt solution volume of 9.9E+07 gallons (SRR-LWP-2009-00001) and the soluble inventory of 0.122 curies. [CBU-PIT-2005-00263]

U-236

The activity concentration for U-236 was determined first by a detection value, then a detection limit. If a detection limit was not reported, the activity concentration was assumed to be 1.1 pCi/mL based on calculations using a total expected salt solution volume of 9.9E+07 gallons (SRR-LWP-2009-00001) and the soluble inventory of 0.406. [CBU-PIT-2005-00263]

U-238

The activity concentration for U-238 was determined first by a detection value, then a detection limit. If a detection limit was not reported, the activity concentration was assumed as 1.0 pCi/mL based on available sample data, detection limits, and time. Note that this assumption is not necessarily applicable to future disposal actions.

Np-237

Np-237 needed no special method. The detection or detection limit value was used.

Pu-238

Pu-238, when no detection or detection limit was available, was assumed to have an activity concentration of 1.0 pCi/mL. This assumption is based on available sample data and detection limits around the same time. Note that this assumption is not necessarily applicable to future disposal actions.

Pu-239

Pu-239, when no detection or detection limit was available, was assumed to have an activity concentration of 1.0 pCi/mL. This assumption is based on available sample data and detection limits around the same time. Note that this assumption is not necessarily applicable to future disposal actions.

Pu-240

Pu-240, when no detection or detection limit was available, was assumed to have an activity concentration of 1.0 pCi/mL. This assumption is based on available sample data and detection limits around the same time. Note that this assumption is not necessarily applicable to future disposal actions.

Pu-241

Pu-241 needed no special method. The detection or detection limit value was used.

Pu-242

Pu-242, when no detection or detection limit was available, was assumed to have an activity concentration of 0.1 pCi/mL. This assumption is based on available sample data and detection limits around the same time. Note that this assumption is not necessarily applicable to future disposal actions.

Pu-244

Pu-244 uses the minimum of detection limit and special method. [CBU-PIT-2005-00039] Dates with no detection limits were populated with the assumed value, 1.00E-03 pCi/mL. This assumption is based on available sample data and detection limits around the same time. Note that this assumption is not necessarily applicable to future disposal actions.

Special method:

where SA_i is the Specific Activity of isotope i

$$\begin{aligned} \text{Mass Pu-244} &= \text{Mass Pu-242} \\ \therefore \text{Pu-244 Activity Concentration} &= \left(\frac{\text{Pu-242 Activity Concentration}}{SA_{\text{Pu-242}}} \right) SA_{\text{Pu-244}} \\ SA_{\text{Pu-242}} &= 3.96 \times 10^{+09} \text{ pCi/g} \\ SA_{\text{Pu-244}} &= 1.83 \times 10^{+07} \text{ pCi/g} \end{aligned}$$

Am-241

Am-241, when no detection or detection limit was available, was assumed to have an activity concentration of 0.1 pCi/mL. This assumed value was based on available sample data, detection limits, and detection limits around the same time. Note that this assumption is not necessarily applicable to future disposal actions.

Am-242m

Am-242m, when no detection or detection limit was available, was assumed to have an activity concentration of 1.0E-02 pCi/mL. This assumption is based on available sample data and detection limits around the same time. Based on the behavior of Am-241 and Am-243, which both increase by approximately two orders of magnitude in activity concentration after the beginning of DDA, it is reasonable to assume such a value for the earlier unknown activity concentrations of Am-242m. Note that this assumption is not necessarily applicable to future disposal actions.

Am-243

Am-243, when no detection or detection limit was available, was assumed to have an activity concentration of 0.19 pCi/mL based on calculations using a total expected salt solution volume of 9.9E+07 gallons (SRR-LWP-2009-00001) and the soluble inventory of 0.07. [CBU-PIT-2005-00263]

Cm-242

Cm-242 is the granddaughter of Am-242m, so the special method activity concentration for Cm-242 was derived from the Am-242m activity concentration. The minimum of the detection limit and special method was used. When no Am-242m detection or detection limit was available, Cm-242 was assumed as 1.0E-02 pCi/mL similarly to Am-242m.

$$\text{Cm-242 Activity Concentration} = 8.2 \times 10^{-01} * \text{Am-242m Activity Concentration}$$

Cm-243

All data for Cm-243 was assumed. The activity concentration was assumed 0.11 pCi/mL using a total expected salt solution volume of 9.9E+07 gallons (SRR-LWP-2009-00001) and a soluble inventory of 4.07E-02 curies. [CBU-PIT-2005-00263]

Cm-244

Cm-244, when no detection or detection limit is available, was assumed to have an activity concentration of 1.0 pCi/mL. This assumption is based on available sample data and detection limits around the same time. Note that this assumption is not necessarily applicable to future disposal actions.

Cm-245

Cm-245 was assumed to have an activity concentration of 3.6E-02 pCi/mL, based on calculations using a total expected salt solution volume of 9.9E+07 gallons (SRR-LWP-2009-00001) and the soluble inventory of 1.34E-02 curies. [CBU-PIT-2005-00263] Recent analysis does provide detection limits, however these are so far above any expected values that a conservative assumption based on total inventory was more reasonable.

Cm-247

The activity concentration for Cm-247 was assumed to be 2.1E-11 pCi/mL based on Table 6 of CBU-PIT-2005-00263, using a soluble inventory of 7.84E-12 curies and a total expected salt solution volume of 9.9E+07 gallons (SRR-LWP-2009-00001). Recent analysis does provide detection limits, however these are so far above any expected values that a conservative assumption based on total inventory was more reasonable.

Cm-248

All data for Cm-248 was assumed. The activity concentration was assumed to be 2.2E-11 pCi/mL based on Table 6 of CBU-PIT-2005-00263, using a soluble inventory of 8.17E-12 curies and a total expected salt solution volume of 9.9E+07 gallons (SRR-LWP-2009-00001). Recent analysis does provide detection limits, however these are so far above any expected values that a conservative assumption based on total inventory was more reasonable.

Bk-249

The activity concentration for Bk-249 was assumed to be 1.6E-18 pCi/mL (corrected for decay) based on calculations using a total expected salt solution volume of 9.9E+07 gallons (SRR-LWP-2009-00001) and the soluble inventory of 5.97E-19 curies with a basis date of 10/1/2005 provided in Table 6 of CBU-PIT-2005-00263. Recent analysis does provide detection limits, however these are so far above any expected values that a conservative assumption based on total inventory was more reasonable. The following expression would be used for decay correction with *Y* representing the year.

$$\text{Bk-249 Activity Concentration} = 1.6 \times 10^{-18} e^{-0.768(Y-2005)}$$

Cf-249

The activity concentration for Cf-249 was assumed to be 1.2E-10 pCi/mL based on Table 6 of CBU-PIT-2005-00263, using a soluble inventory of 4.53E-11 curies and a total expected salt solution volume of 9.9E+07 gallons (SRR-LWP-2009-00001). Recent analysis does provide detection limits, however these are so far above any expected values that a conservative assumption based on total inventory was more reasonable.

Cf-251

The activity concentration for Cf-251 was assumed to be 4.13E-12 pCi/mL based on Table 6 of CBU-PIT-2005-00263, using a soluble inventory of 1.55E-12 curies and a total expected salt solution volume of 9.9E+07 gallons (SRR-LWP-2009-00001). A detected concentration reported in Q-CLC-Z-00001 for Cf-251 on 7/17/2003 and recently reported detection limits are not included in this methodology because these values are unreasonable based on the total estimated inventory within the SRS tank farms.

Cf-252

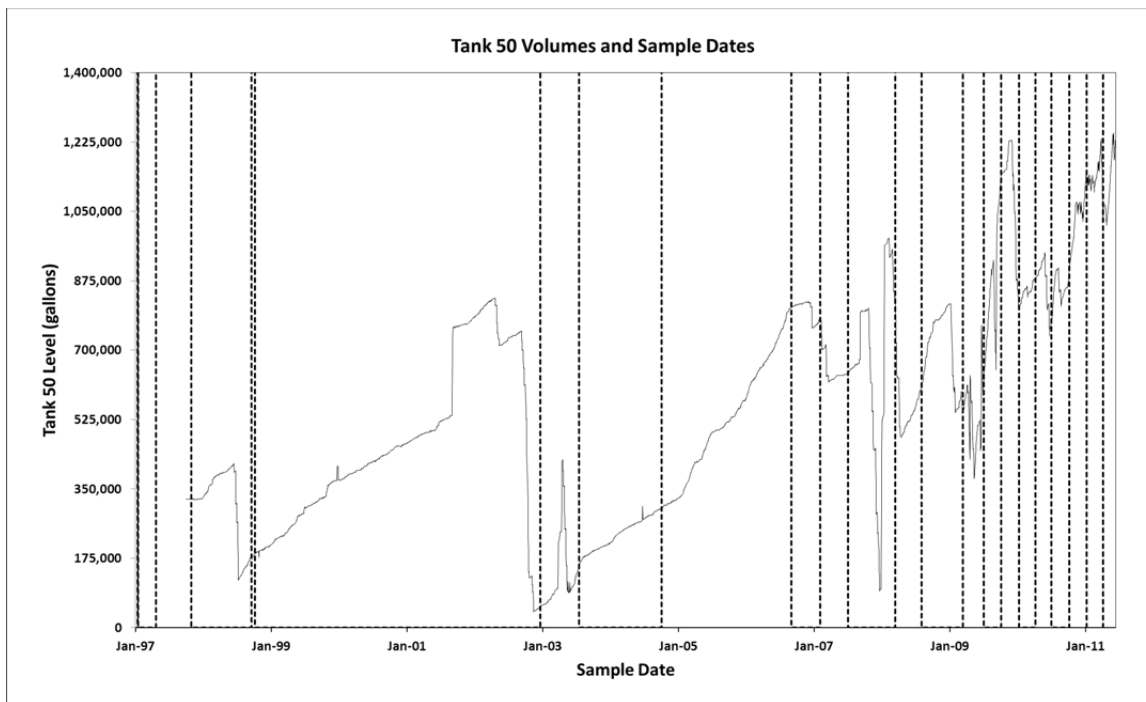
The activity concentration for Cf-252 was determined by the detection limit. If a detection limit was not reported, the activity concentration was assumed to be $1.34\text{E-}13$ pCi/mL (corrected for decay) based on calculations using a total expected salt solution volume of $9.9\text{E+}07$ gallons (SRR-LWP-2009-00001) and the soluble inventory of $5.03\text{E-}14$ curies with a basis date of 10/1/2005 provided in Table 6 of CBU-PIT-2005-00263. Decay was included due to the short (2.65 years) half-life of Cf-252. Recent analysis does provide detection limits, however these are so far above any expected values that a conservative assumption based on total inventory was more reasonable. The following expression was used for decay correction with Y as the year:

$$\text{Cf-252 Activity Concentration} = 1.34 \times 10^{-13} e^{-0.262(Y-2005)}$$

3.3 Corresponding Sample and Transfer Data

Once complete sample information was compiled for each sample date, a sample was assigned to each transfer to represent that material. Sample information (activity concentrations by radionuclide) for Tank 50 is currently reported on a quarterly basis, although historically there were gaps where samples were not taken and/or processing did not occur, as shown in Figure 3.3-1. Samples are shown in the figure as vertical dashed lines, while the Tank 50 volume is shown by the solid black line.

Figure 3.3-1: Tank 50 Volumes and Sample Dates



Transfers from Tank 50 were methodically represented by the most recent sample, with one sample representing all transfers until the next sample was taken. If a sample was taken in the middle of a quarter, the transfers before the sample date use the previous sample and transfers on the day of the sample and later use the new sample. All samples represent only material processed after the date of the sample, except for the first sample, which was taken after

processing began. In the case of Vault 1, where the transfer volumes are available by month, this was performed to the applicable degree (e.g., a sample taken 4/26/1996 began representing material processed beginning in May of 1996). For the one date with multiple samples, the average of the concentrations was used to represent corresponding transfers. The periods of processing for Vaults 1 and 4 (by calendar quarter or month) are shown in Table 3.3-1 and Table 3.3-2, with the dates of the samples used to represent the processed material, the concentration reference, and the processing volume reference. All transfers reported by month were considered to have occurred on the first day of the month for purposes of matching them with a representative sample.

Table 3.3-1: Transfer Periods and Sample Dates for Vault 1

<i>Processing Period</i>	<i>Sample Date</i>	<i>Activity Concentration Reference</i>	<i>Processing Volume Reference</i>
Q3 1996	4/26/1996	Q-CLC-Z-00001	WSRC-RP-2008-00390
Jun-1996	4/26/1996	Q-CLC-Z-00001	WSRC-RP-2008-00390
May-1996	4/26/1996	Q-CLC-Z-00001	WSRC-RP-2008-00390
Apr-1996	9/19/1995	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q1 1996	9/19/1995	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q4 1995	9/19/1995	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q3 1995	2/10/1995	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q2 1995	2/10/1995	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q1 1995	10/6/1994	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q4 1994 ^a	10/6/1994	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q4 1994	6/9/1994	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q3 1994	6/9/1994	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q2 1994	2/15/1994	Q-CLC-Z-00001	WSRC-RP-2008-00390
Mar-1994	2/15/1994	Q-CLC-Z-00001	WSRC-RP-2008-00390
Feb-1994	8/13/1992	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q4 1993	8/13/1992	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q3 1993	8/13/1992	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q1 1993	8/13/1992	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q4 1992	8/13/1992	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q2 1992	1/29/1992	Q-CLC-Z-00001	WSRC-RP-2008-00390
Feb -1992	1/29/1992	Q-CLC-Z-00001	WSRC-RP-2008-00390
Jan-1992	7/15/1991	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q4 1991	7/15/1991	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q3 1991	7/15/1991	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q2 1991	7/25/1990	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q1 1991	7/25/1990	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q4 1990	7/25/1990	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q3 1990	7/25/1990	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q2 1990 ^b	7/25/1990	Q-CLC-Z-00001	WSRC-RP-2008-00390

^a Transfers after 10/5/1994

^b Includes transfers prior to 7/25/1990

Table 3.3-2: Transfer Periods and Sample Dates for Vault 4

<i>Processing Period</i>	<i>Sample Date</i>	<i>Activity Concentration Reference</i>	<i>Processing Volume Reference</i>
Q3 2011	7/7/2011	SRNL-STI-2011-00574	X-CLC-Z-00042
Q2 2011 ^a	4/4/2011	SRNL-STI-2011-00443	X-CLC-Z-00041
Q2 2011	1/5/2011	SRNL-STI-2011-00303	X-CLC-Z-00041
Q1 2011	1/5/2011	SRNL-STI-2011-00303	X-CLC-Z-00038
Q4 2010	10/4/2010	SRNL-STI-2010-00713	X-CLC-Z-00035
Q3 2010	7/1/2010	SRNL-STI-2010-00598	X-CLC-Z-00032
Q2 2010	4/5/2010	SRNL-STI-2010-00437	SRR-WSE-2010-00162
Feb-2010	1/8/2010	SRNL-STI-2010-00210	SRR-WSE-2010-00076
Jan-2010	10/2/2009	SRNL-STI-2009-00828	SRR-WSE-2010-00076
Q4 2009	10/2/2009	SRNL-STI-2009-00828	SRR-WSE-2010-00011
Q3 2009	7/1/2009	SRNL-STI-2009-00585	SRR-WSE-2009-00090
Q2 2009	3/10/2009	SRNL-STI-2009-00452	SRR-WSE-2009-00019
Q1 2009 ^c	3/10/2009	SRNL-STI-2009-00452	LWO-WSE-2009-00095
Q1 2009	7/31/2008	SRNS-TR-2008-00328	LWO-WSE-2009-00095
Q2 2008	3/11/2008	WSRC-TR-2008-00184	LWO-WSE-2008-00135
Q1 2008 ^d	3/11/2008	WSRC-TR-2008-00184	LWO-WSE-2008-00067
Q1 2008	9/1/2007 ^b	WSRC-TR-2008-00184	LWO-WSE-2008-00067
Q4 2007	9/1/2007 ^b	WSRC-TR-2008-00080	LWO-WSE-2008-00012
Q1 2007	2/1/2007 ^b	WSRC-TR-2007-00253	WSRC-RP-2008-00390
Q4 2006	9/1/2006 ^b	WSRC-TR-2007-00133	WSRC-RP-2008-00390
Q3 2003	7/17/2003	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q2 2003	12/20/2002	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q4 2002	10/5/1998	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q3 2002	10/5/1998	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q2 2002	10/5/1998	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q3 1998	10/28/1997	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q2 1998	10/28/1997	Q-CLC-Z-00001	WSRC-RP-2008-00390
Q1 1998	10/28/1997	Q-CLC-Z-00001	WSRC-RP-2008-00390
Dec-1997	10/28/1997	Q-CLC-Z-00001	WSRC-RP-2008-00390
Nov-1997	10/28/1997	Q-CLC-Z-00001	WSRC-RP-2008-00390
Oct-1997	4/23/1997 ^e	Q-CLC-Z-00001	WSRC-RP-2008-00390
Jun-1997	4/23/1997 ^e	Q-CLC-Z-00001	WSRC-RP-2008-00390
May-1997	4/23/1997 ^e	Q-CLC-Z-00001	WSRC-RP-2008-00390
Apr-1997	1/15/1997	Q-CLC-Z-00001	WSRC-RP-2008-00390
Mar-1997	1/15/1997	Q-CLC-Z-00001	WSRC-RP-2008-00390
Feb-1997	1/15/1997	Q-CLC-Z-00001	WSRC-RP-2008-00390
Jan-1997	9/20/1996	Q-CLC-Z-00001	WSRC-RP-2008-00390

^a Transfers after 4/3/2011

^b No exact sample date given in the report, so the sample date was assumed to be taken on the first of the month.

^c Transfers after 3/9/2009

^d Transfers after 3/10/2008

^e Two samples were taken on this date. The activity concentrations were averaged by radionuclide to provide the representative sample used in the inventory calculations.

3.4 Decay Calculations

Decay was performed from the middle of the original quarter of disposal (e.g., inventory from the second calendar quarter of 2009 was decayed from 5/15/2009) to the end of FY2011, or 9/30/2011. All decay calculations were performed in ORIGEN 2.2 Isotope Generation and Depletion Code using the ORIGEN2_for_CWDA_Users_v1.0.xlsm file (SRR-CWDA-2011-000146) to interpret the input data into the input text file required by ORIGEN. ORIGEN reports output data by type of product, so some radionuclides show up more than once. For this effort, these values were added together to produce the total inventory reported here.

In order to compensate for very low initial inventories of certain radionuclides such as Bk-249, the entire inventory was increased by 16 orders of magnitude, decayed, and then reduced by 16 orders of magnitude. This compensated for ORIGEN's inability to interpret low initial inventories while maintaining the entire data set for decay and ingrowth.

3.5 Updated Inventories

Using the methods described previously, updated inventories for Vault 1 and 4 were calculated and decayed from the original quarter of disposal to 9/30/2011. The resulting inventory for each radionuclide was summed over all of the quarters of disposal to provide a total inventory for each SDU. These inventories are presented in Table 3.5-1 and Table 3.5-2.

Table 3.5-1: Updated Vault 1 Inventory

Rad	Ci	Rad	Ci	Rad	Ci	Rad	Ci
H-3	1.9E+01	Pd-107	8.4E-03	Pt-193	1.8E+00	Pu-241	2.7E-02
C-14	1.3E+00	Sn-126	1.2E+00	Ra-226	4.2E-07	Pu-242	1.6E-03
Na-22	8.6E+00	Sb-125	1.1E-01	Ra-228	7.7E-06	Pu-244	1.0E-05
Al-26	2.6E-01	Sb-126	1.7E-01	Ac-227	1.4E-06	Am-241	1.7E-03
Cl-36	1.9E-07	Sb-126m	1.2E+00	Th-229	3.6E-04	Am-242m	7.0E-05
K-40	1.9E-07	Te-125m	2.6E-02	Th-230	4.7E-05	Am-243	1.4E-03
Co-60	9.9E-04	I-129	2.0E-01	Th-232	7.7E-06	Cm-242	5.8E-05
Ni-59	2.3E-03	Cs-134	7.5E-03	Pa-231	2.7E-06	Cm-243	5.4E-04
Ni-63	1.2E-01	Cs-135	9.8E-02	U-232	7.0E-04	Cm-244	3.8E-03
Se-79	3.4E-01	Cs-137	7.3E+00	U-233	7.8E-02	Cm-245	2.8E-04
Sr-90	1.3E-02	Ba-137m	6.9E+00	U-234	1.0E-01	Cm-247	1.6E-13
Y-90	1.3E-02	Ce-144	6.4E-02	U-235	2.5E-03	Cm-248	1.7E-13
Zr-93	7.3E-01	Pr-144	6.3E-02	U-236	6.5E-03	Bk-249	1.4E-22
Nb-93m	7.3E-01	Pm-147	1.6E-02	U-238	1.1E-02	Cf-249	9.0E-13
Nb-94	2.0E-03	Sm-151	4.9E-03	Np-237	3.9E-03	Cf-251	3.1E-14
Tc-99	5.5E+01	Eu-152	2.1E-03	Pu-238	1.0E-02	Cf-252	2.2E-16
Ru-106	2.7E-05	Eu-154	8.6E-04	Pu-239	1.8E-02		
Rh-106	2.7E-05	Eu-155	6.8E-04	Pu-240	1.7E-02	Total	1.1E+02

Table 3.5-2: Updated Vault 4 Inventory

Rad	Ci	Rad	Ci	Rad	Ci	Rad	Ci
H-3	3.3E+01	Pd-107	3.2E-02	Pt-193	8.3E+00	Pu-241	1.2E+02
C-14	5.8E+00	Sn-126	1.8E+00	Ra-226	2.5E-05	Pu-242	4.0E+00
Na-22	2.4E+01	Sb-125	1.2E+02	Ra-228	2.0E-04	Pu-244	1.6E-02
Al-26	9.1E-01	Sb-126	2.6E-01	Ac-227	1.8E-05	Am-241	1.9E+01
Cl-36	2.9E-02	Sb-126m	1.8E+00	Th-229	4.0E-02	Am-242m	1.9E-02
K-40	2.9E-02	Te-125m	3.0E+01	Th-230	2.5E-03	Am-243	5.0E-01
Co-60	1.3E-01	I-129	2.6E-01	Th-232	2.0E-04	Cm-242	1.6E-02
Ni-59	8.0E-02	Cs-134	1.1E+01	Pa-231	3.5E-05	Cm-243	2.9E-03
Ni-63	3.2E+00	Cs-135	1.7E+00	U-232	1.2E-01	Cm-244	4.0E+01
Se-79	9.7E+00	Cs-137	1.9E+05	U-233	8.4E+00	Cm-245	1.0E-03
Sr-90	2.7E+03	Ba-137m	1.8E+05	U-234	5.4E+00	Cm-247	6.1E-13
Y-90	2.7E+03	Ce-144	1.0E+00	U-235	3.3E-02	Cm-248	6.4E-13
Zr-93	7.3E+00	Pr-144	1.0E+00	U-236	7.7E-02	Bk-249	1.6E-21
Nb-93m	7.6E+00	Pm-147	1.4E+01	U-238	6.6E-02	Cf-249	3.5E-12
Nb-94	8.9E-02	Sm-151	1.7E+01	Np-237	5.1E-01	Cf-251	1.2E-13
Tc-99	5.3E+02	Eu-152	1.0E-01	Pu-238	3.2E+02	Cf-252	9.8E-16
Ru-106	1.9E-01	Eu-154	5.0E+00	Pu-239	5.8E+01		
Rh-106	1.9E-01	Eu-155	1.6E+00	Pu-240	7.2E+01	Total	3.7E+05

4.0 CONCLUSIONS

As previously stated, earlier radiological inventories for the SDUs known as Vault 1 and Vault 4 have employed conservative assumptions concerning certain radionuclides entering Tank 50, the SPF feed waste tank. This effort provides a more methodical approach that is used to recalculate the inventory of both SDUs from the original Tank 50 sample reports and transfer information. The combination of decay from the original quarter of disposal and reduction of conservative historical assumptions results in a total Vault 1 inventory of 0.1 kilocuries and a total Vault 4 inventory of 374 kilocuries.

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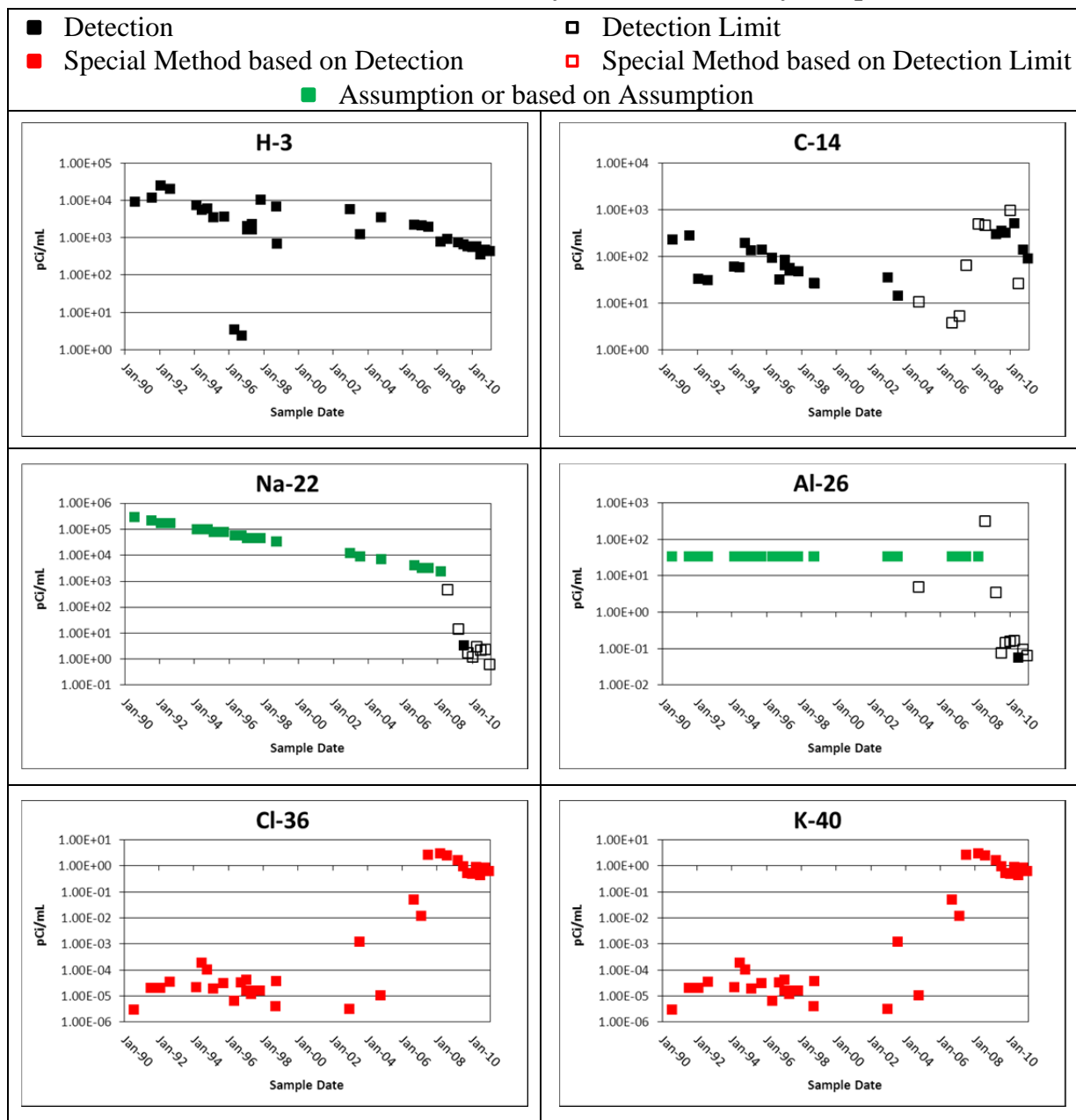
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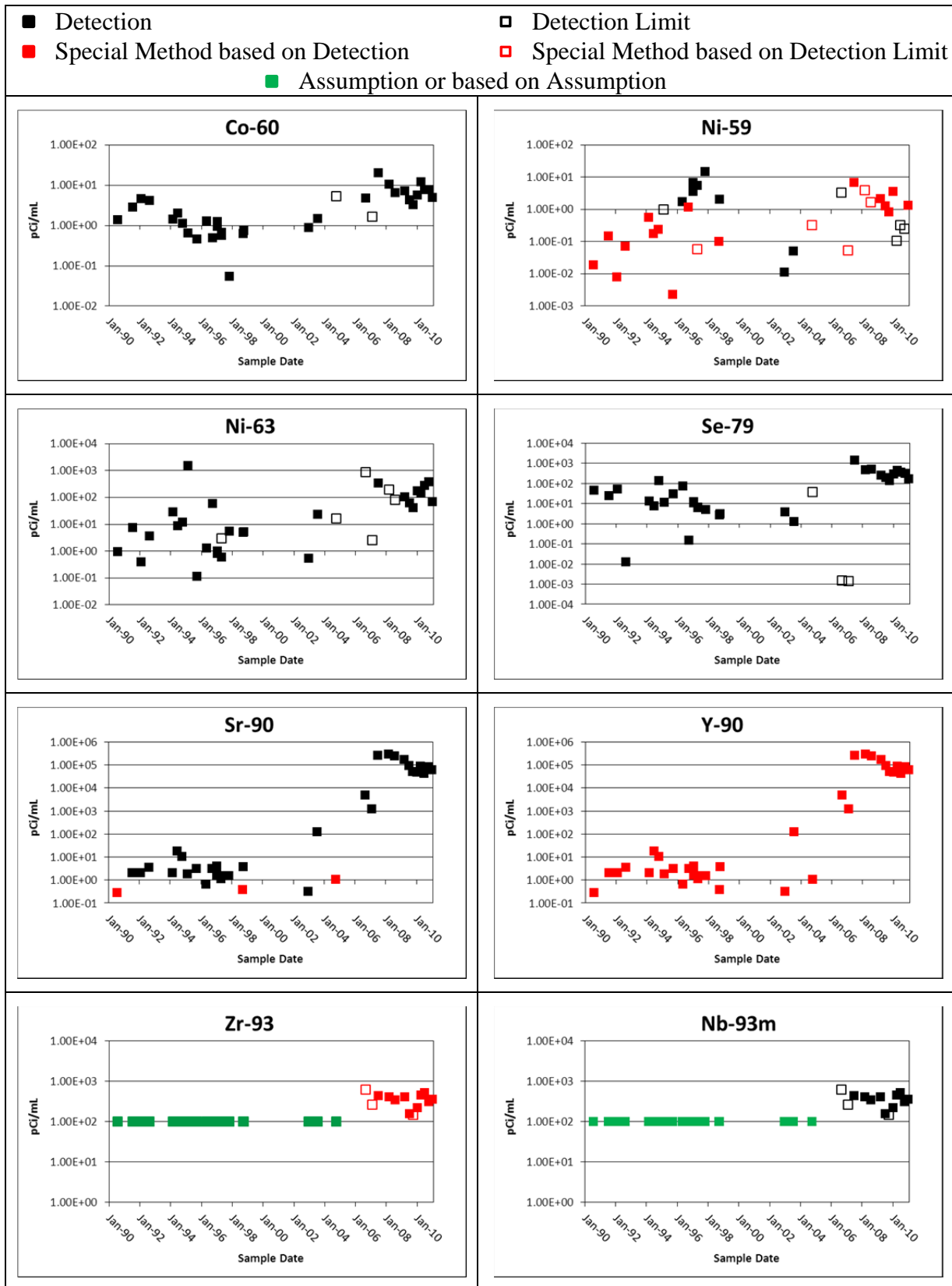
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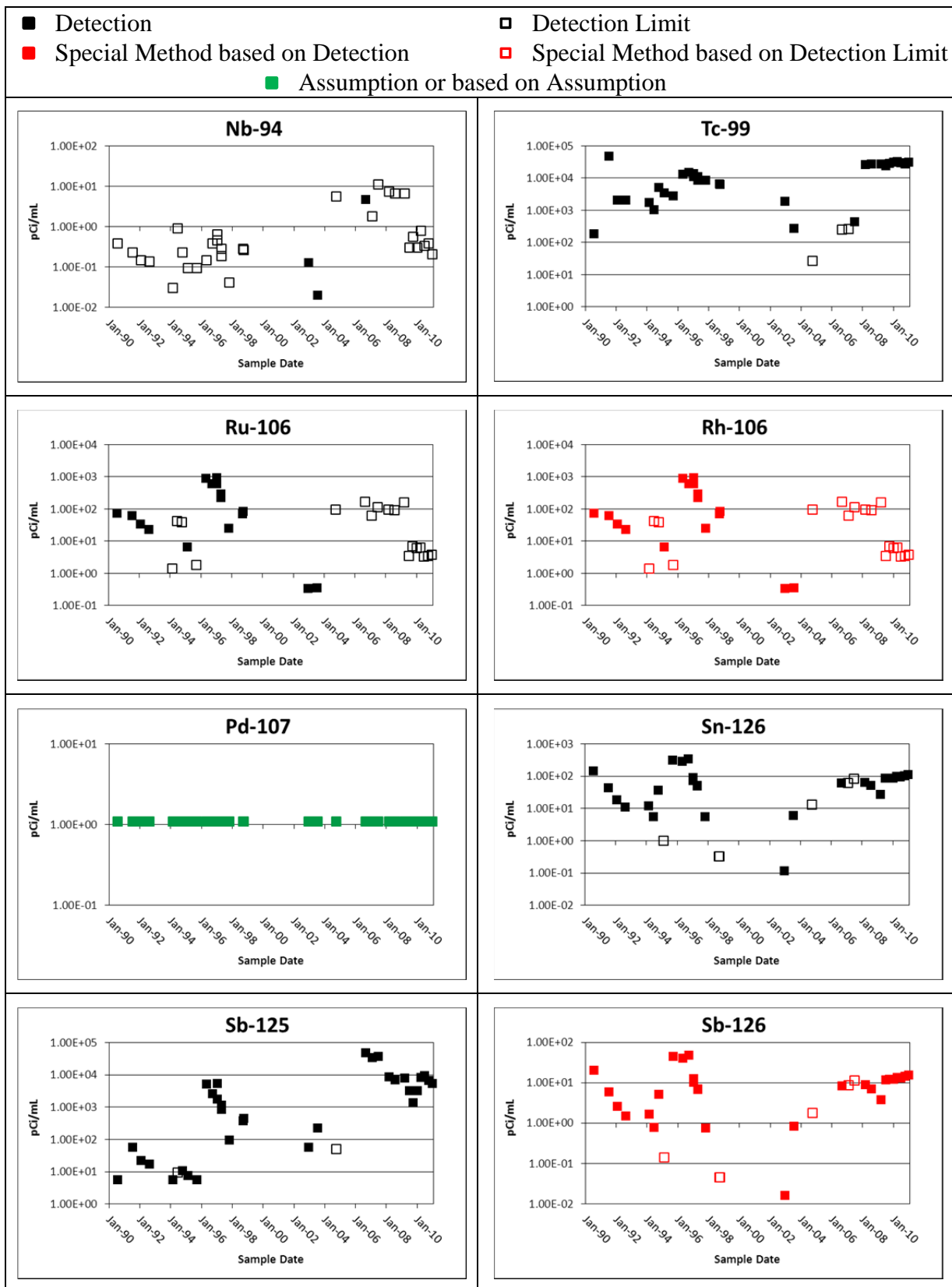
APPENDIX A

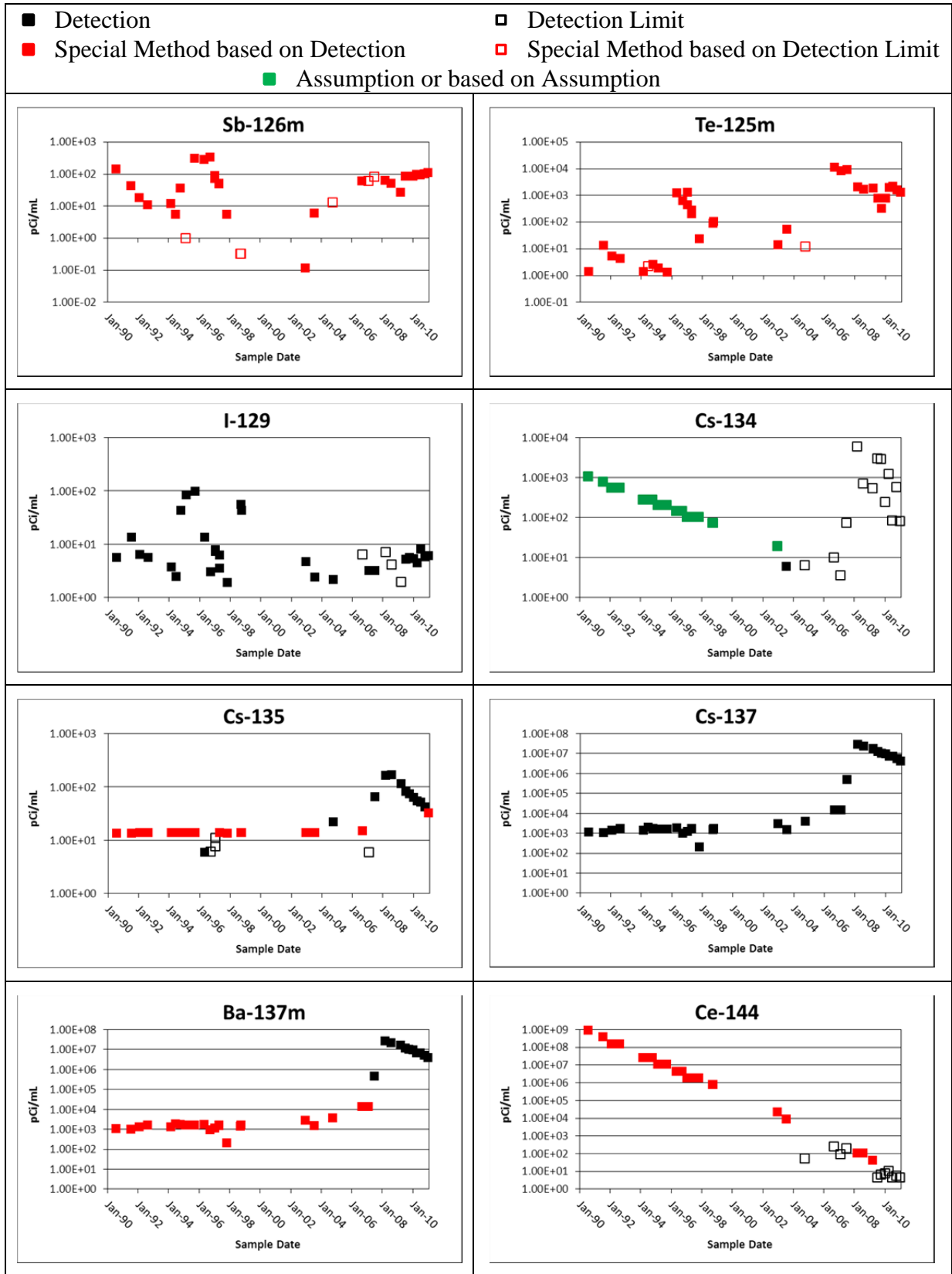
Presented here in Table A.0-1 are the activity concentrations over time used for each radionuclide according to the special methods presented in Section 3.3. Each square represents an activity concentration, with solid black representing detections, open black representing detection limits, solid red representing a special method calculation based on a detection, open red representing a special method calculation based on a detection limit, and solid green representing an assumption or a decay calculation based on an assumed value.

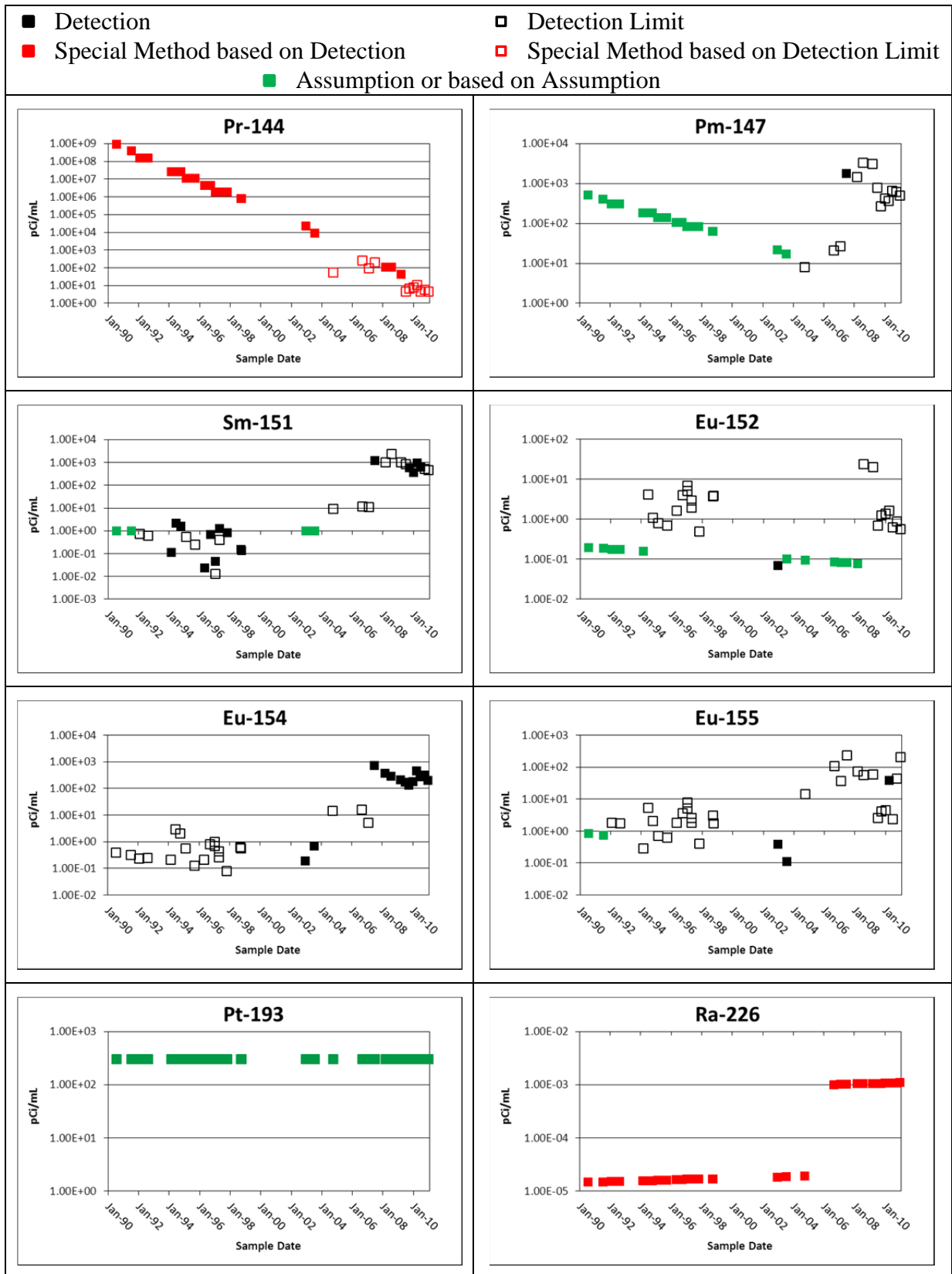
Table A.0-1: Radionuclide Activity Concentrations by Sample Date

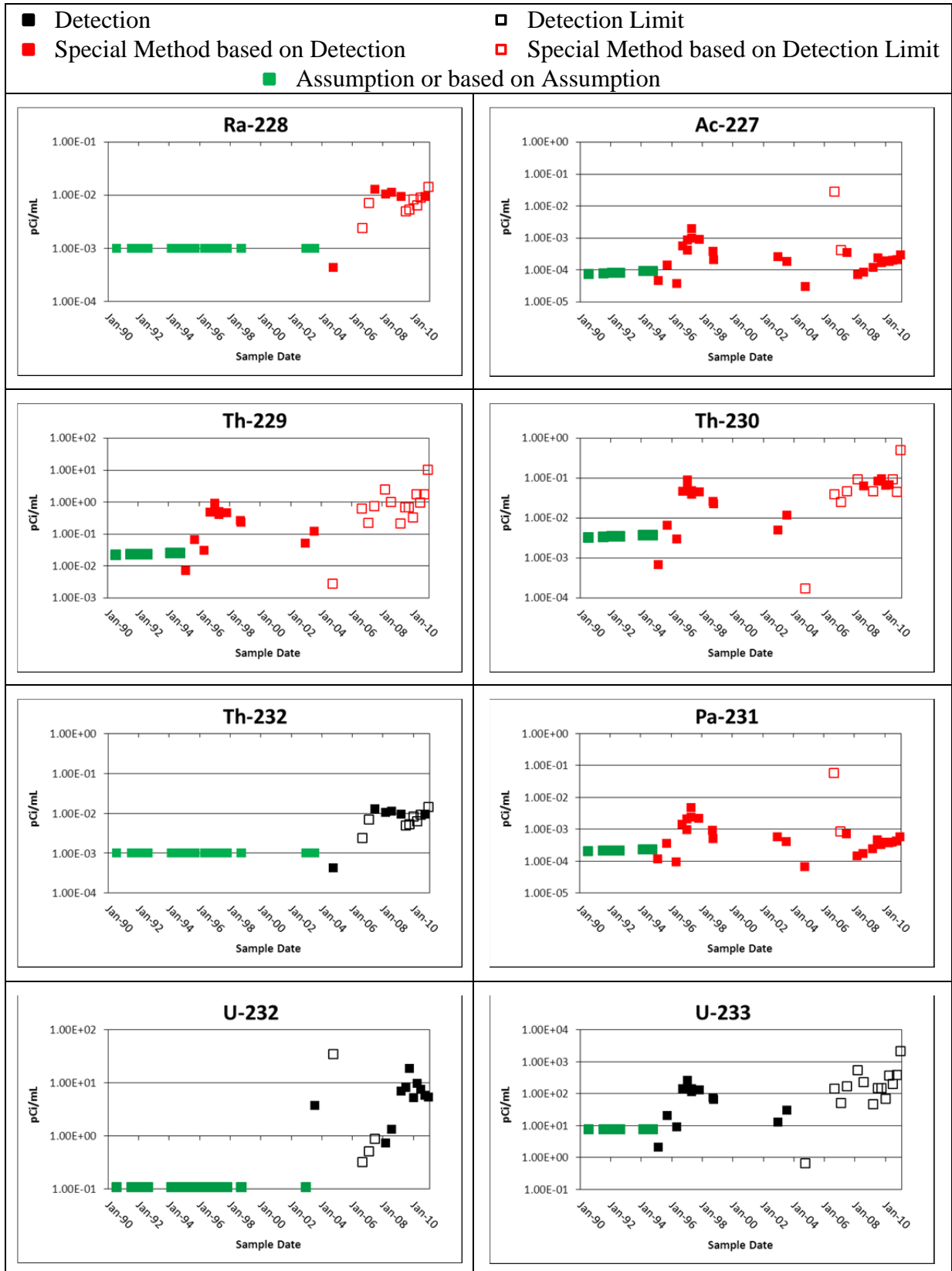


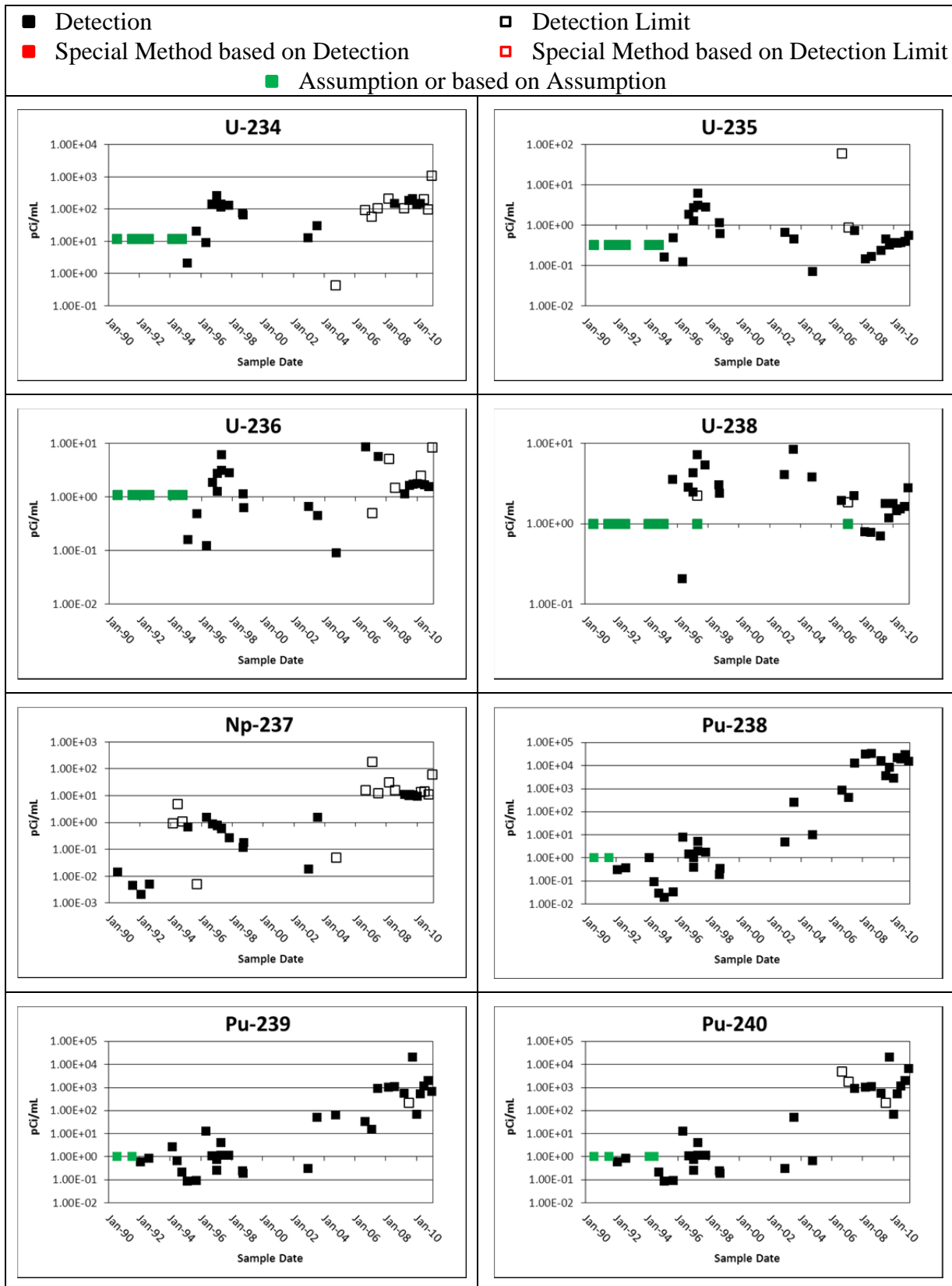


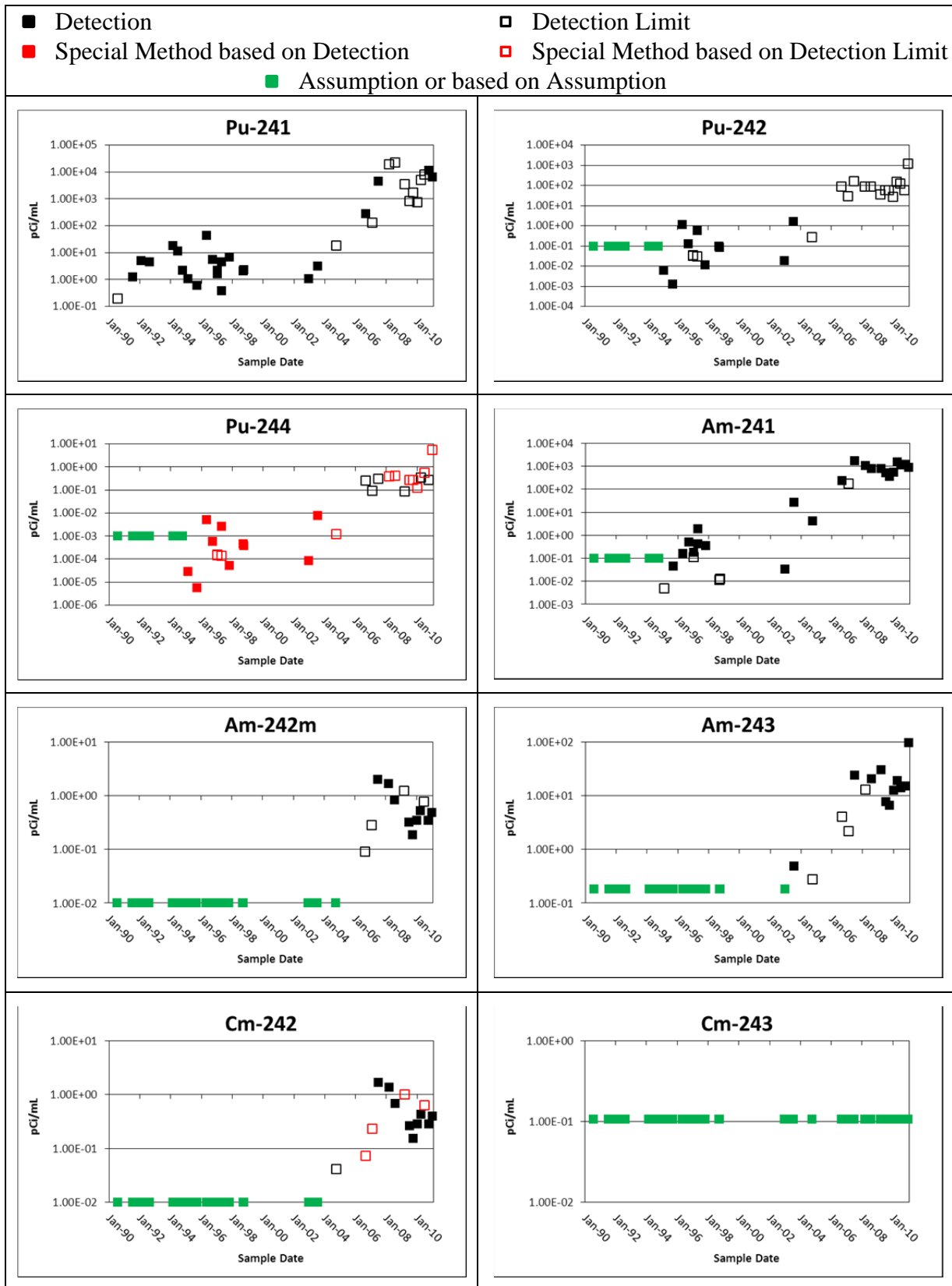


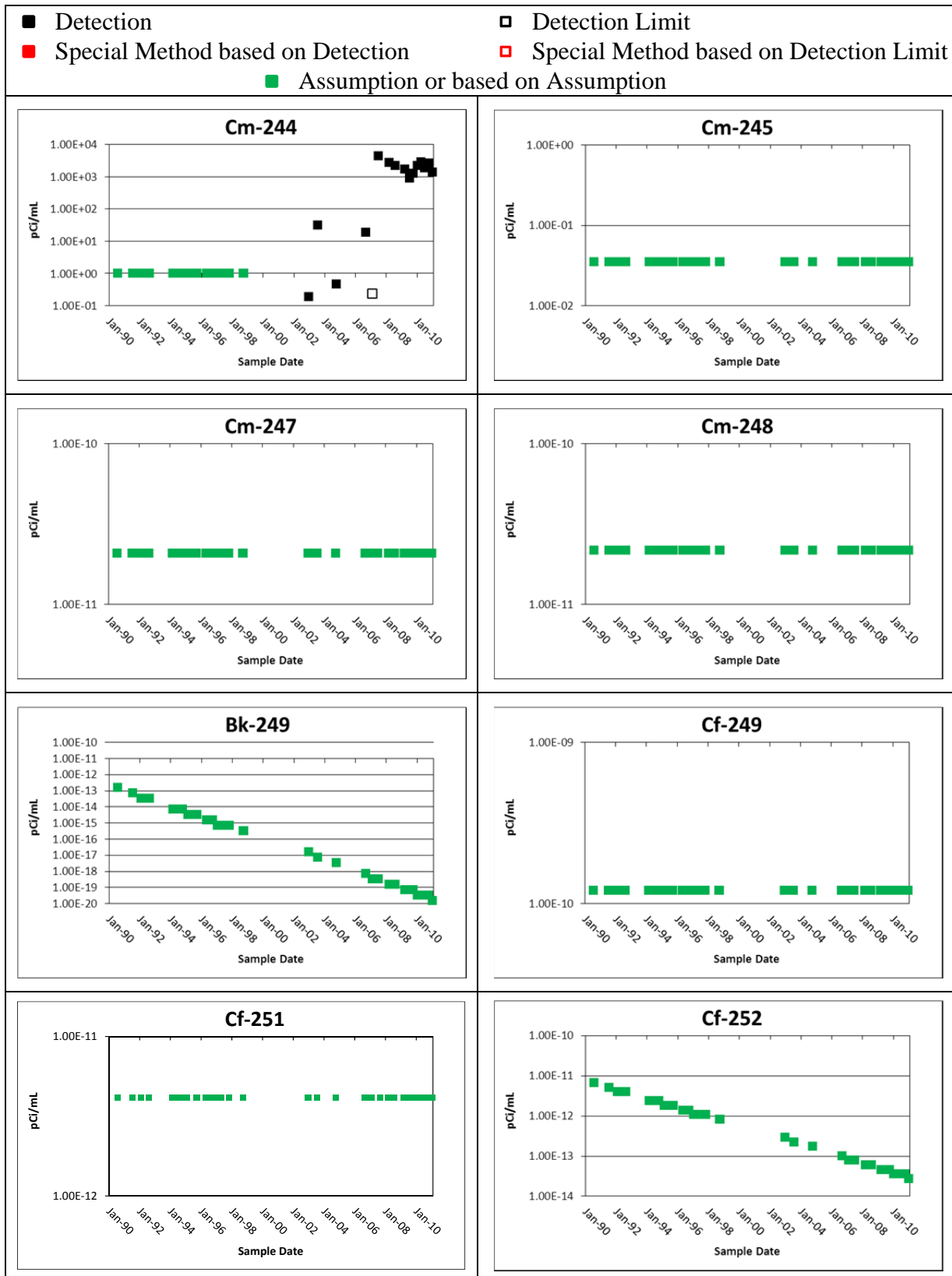












APPENDIX B

A regression analysis was used to estimate the Cs-135 concentration in Tank 50 when its detection was not reported. Table B.0-1 presents the sampling data when both Cs-135 and Cs-137 concentrations were reported from the Tank 50 sample analysis reports issued as of June 2011. Sampling reports that reported detection limits rather than actual detected concentrations are not included in Table B.0-1. Also included in Table B.0-1 is the Cs-137 to Cs-135 concentration ratios.

Table B.0-1: Cs-135 and Cs-137 Reported Concentrations in pCi/mL

Reference	Sample Date	Cs-135	Cs-137	Ratio
SRNL-STI-2011-00443	4/4/2011	1.94E+01	3.28E+06	1.69E+05
SRNL-STI-2010-00713	10/4/2010	4.22E+01	5.54E+06	1.31E+05
SRNL-STI-2010-00598	7/1/2010	5.21E+01	7.31E+06	1.40E+05
SRNL-STI-2010-00437	4/5/2010	5.43E+01	7.37E+06	1.36E+05
SRNL-STI-2010-00210	1/8/2010	6.37E+01	9.79E+06	1.54E+05
SRNL-STI-2009-00828	10/2/2009	7.35E+01	1.01E+07	1.37E+05
SRNL-STI-2009-00585	7/1/2009	8.20E+01	1.26E+07	1.54E+05
SRNL-STI-2009-00452	3/10/2009	1.16E+02	1.73E+07	1.49E+05
SRNS-TR-2008-00328	7/31/2008	1.68E+02	2.31E+07	1.38E+05
WSRC-TR-2008-00184	3/11/2008	1.65E+02	2.86E+07	1.73E+05
WSRC-TR-2008-00080	9/1/2007	6.45E+01	5.02E+05	7.78E+03
WSRC-TR-2004-00625	10/1/2004	2.19E+01	4.04E+03	1.84E+02
Q-CLC-Z-00001	4/26/1996	6.02E+00	1.89E+03	3.14E+02

Inspection of the ratios presented in Table B.0-1 indicates that the regression analysis should be conducted for two periods of time, prior to and after 03/11/2008.

Using the regression analysis tool from Microsoft Excel for a linear fit of the data (LINEST), the following regression analyses were generated.

Prior to 03/11/2008:

$$\text{Cs-135 Activity Concentration (pCi/mL)} = 1.0 \times 10^{-04} * \text{Cs-137 Activity Concentration} + 13.6$$

03/11/2008 and forward:

$$\text{Cs-135 Activity Concentration (pCi/mL)} = 6.11 \times 10^{-06} * \text{Cs-137 Activity Concentration} + 7.24$$