

ESTIMATION OF Ni⁶³, Pu²⁴¹, Pu²⁴² AND Am²⁴³ FROM Co⁶⁰, Pu²³⁹, AND Cm²⁴⁴ ACTIVITIES IN GROUNDWATER SAMPLES

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A document prepared for:
requested by Environmental Restoration Department Subcontractors
at
from thru

DOE Contract No. DE-AC09-89SR18035

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WESTINGHOUSE SAVANNAH RIVER COMPANY
INTER-OFFICE MEMORANDUM



WSRC-RP-93-0723

May 14, 1993

TO: C. M. LEWIS, 992-4W

FROM: H. P. HOLCOMB, 992-4W (644-6745)

HPH

ESTIMATION OF Ni⁶³, Pu²⁴¹, Pu²⁴², AND Am²⁴³ FROM Co⁶⁰,
Pu²³⁹, AND Cm²⁴⁴ ACTIVITIES IN GROUNDWATER SAMPLES (U)

INTRODUCTION AND SUMMARY

The Part B Permit for F&H Seepage Basins calls for analysis of several constituents of concern in groundwater monitoring wells. Four of these analytes are the radionuclides Ni⁶³, Pu²⁴¹, Pu²⁴², and Am²⁴³. These are currently not being analyzed due to their very difficult, tedious analytical schemes coupled with their relatively low activity values.

WSRC has requested SCDHEC to grant relief from monitoring these four radionuclides (rads) as specified in the Part B Permit. If relief is denied, the option of estimating these four rads is outlined in this report that could serve as the technical basis for proposing this option to SCDHEC. This approach offers a very cost-effective manner by which to monitor these analytes by means of estimating their activities via correlation with those of more easily analyzed, chemically-similar rads.

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This report demonstrates how the activity value for Ni^{63} , a weak beta emitter, can be estimated from that of Co^{60} , an easily detectable, high-energy gamma emitter. Similarly, estimates of Pu^{241} , a beta emitter, and the alpha-emitting Pu^{242} can be made from the activity value of the more easily detected Pu^{239} . Am^{243} can be estimated from the activity of Cm^{244} , which is easier to detect because of a shorter half-life (higher specific activity) and the emission of higher energy alpha particles.

These correlations are made under very specific parameters in order to ensure the validity of this approach. Therefore, assumptions must be established setting ground rules for establishing these activity relationships. Bases for these assumptions are explained and/or referenced. Their degree of uncertainty limits the accuracy of the data so that the term "estimate" is used. It is difficult to assign error bars to the estimates, but I believe they are within factors of $\pm 2x$ to $\pm 3x$.

Such soundly-based, conservative estimates for these four radcs can provide a tool for evaluating any hazards from their presence over the next several years. Hopefully, during this time, sufficient advances will be made in their radiochemical analyses and in counting techniques so that in the future, their activities may be quantitatively determined more easily and also more cost effectively.

For the present, I plan to evaluate further this correlational technique to determine similar activity relationships between other rad analytes and to ascertain if certain analyses can be eliminated (as cost savings) without adversely affecting needed monitoring information.

DISCUSSION

Correlating Ni^{63} with Co^{60}

Ni^{63} is an activation product produced in SRS reactors from the neutron capture by the Ni^{62} isotope, one of five stable nickel isotopes present in stainless steel components. Similarly, Co^{60} is produced by neutron capture by the only stable isotope of cobalt, Co^{59} , which is present as an impurity in the stainless steel.

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N. P. Baumann of the Scientific Computations Section of SRTC was requested¹ to calculate the Ni^{63}/Co^{60} activity ratio at reactor shutdown following irradiation. For calculational bases furnished to Baumann, contacts were made with H. B. Peacock, Jr.², of the Materials Technology Section of SRTC and with Bob Brown³ of Carpenter Technologies, a speciality steel company and previous supplier of stainless steel reactor components to SRS. Peacock indicated that the stainless steel used in SRS reactors was either 304 or 304L, the latter differing from the former by having a lower carbon content. He also furnished analyses by Slater Steels Corporation showing the cobalt content of 304L furnished SRS in 1991 to be 0.09 wt % and nickel, 9.97 wt %. Brown indicated there were two current specifications for cobalt in 304/304L used in nuclear applications: 0.1 wt % maximum or 0.2 wt % maximum.

For conservatism, Baumann chose 0.1 wt % for the cobalt content (to yield a higher Ni^{63}/Co^{60} value) and 9.5 wt % for the nickel content of 304/304L reactor components. His bases and results⁴ indicate that the Ni^{63}/Co^{60} activity ratio immediately following irradiation of stainless steel with the composition described above is 0.05723.

In applying this ratio to the activities in groundwater other assumptions must be made. These are:

1. Migratory Ni^{63} and Co^{60} in the samples came from a charge in an SRS reactor in which irradiation ceased on Jan. 1, 1971. This date is chosen because this is approximately midway during the production mode operating period (1954-1988) for SRS reactors during most of which the F and H Seepage Basins were active.
2. No separation of the Ni^{63} and Co^{60} occurred during processing or during environmental residence. Both elements in solution are principally divalent. Cobalt can exist as the +3 ion, but it is such a powerful oxidizing agent that it can only exist in the higher oxidation state in a sequestered, chemically-pristine environment. Even then, it can decompose water thereby being reduced to the +2 ion.⁵ The +2 ions of cobalt and nickel exhibit similar chemistry. They are adjacent members of the first series of transition metals in which the 3d electron orbitals are being filled. Both form stable ammonia complexes, both form hydroxides with similar K_{sp} s, and both form relatively insoluble sulfides.⁶ Both behave quite similarly

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in SRS separations and waste management processes because of their chemical likeness.

Based on Baumann's calculated ratio and using the foregoing assumptions, the information in Table 1 can be derived to yield activity ratios at different times for these two rads. The standard decay equation, $n = n_0 e^{-\lambda t}$ [where "n" = the number of atoms or activity at time "t", "n₀" = the number of atoms or activity at time = 0 (end of irradiation), and "λ" = the decay constant (or 0.693/t_{1/2})], was used to calculate the respective fractions remaining via spreadsheet software.

(NOTE: All half-lives used in calculations for this section and the ones following came from the same source and are referenced in the footnotes of each Table.)

Because of the significant difference in half-lives for these two rads, the current activity ratio is approximately 1. In three years, 1996, the activity of the Ni⁶³ will be about 1.3 times that of Co⁶⁰. A plot of the ratio versus time is shown in Figure 1.

The primary limitation for using this particular correlation to estimate Ni⁶³ is that Ni⁶³/Co⁶⁰ is increasing relatively rapidly with time. By the year 2006, or 13 years from now, the ratio will reach 4.5. Therefore, as the Co⁶⁰ decays and its activity level decreases, the point will be reached where possibly significant activity values of Ni⁶³ will exist at activity levels of Co⁶⁰ that are less than detectable. However, for the next decade, the ratio should be reliable and valid for estimating purposes.

Correlating Pu²⁴¹ and Pu²⁴² With Pu²³⁹

There are no chemical or environmental assumptions in this comparison because these are isotopes of the same element and will act the same. No known evidence exists for any separation of plutonium isotopes in SRS processes or in the environment.

Assumptions that are made are:

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1. Similar to the $\text{Ni}^{63}/\text{Co}^{60}$ case above, the migratory Pu^{241} , Pu^{242} , and Pu^{239} in the samples came from a charge in an SRS reactor in which irradiation ceased on Jan. 1, 1971. Again, this date is chosen because this is approximately midway during the production mode operating period (1954-1988) for SRS reactors.

2. Weapons-grade plutonium is considered to be the source term with the following isotopic weight percents⁷ at reactor shutdown: Pu^{238} - 0.007, Pu^{239} - 93.41, Pu^{240} - 5.86, Pu^{241} - 0.70, and Pu^{242} - 0.03. Such an assumption is valid because most of the plutonium produced at SRS has been weapons-grade with nominally 6 wt % or less Pu^{240} .

Using the foregoing assumptions and the standard decay equation, the information shown in Table 2 can be developed. The activity ratios for $\text{Pu}^{241}/\text{Pu}^{239}$ and for $\text{Pu}^{242}/\text{Pu}^{239}$ versus time are plotted in Figure 2. From the data and from the plot it can be seen that the $\text{Pu}^{241}/\text{Pu}^{239}$ activity ratio is currently around 4.2 and decreasing slowly. Around the year 2021, or 28 years from now, the ratio will be approximately 1.

Likewise, the current $\text{Pu}^{242}/\text{Pu}^{239}$ activity ratio is $2\text{E}-05$ and will remain at that level for the next 1000 years at least. This small value indicates the very low hazard produced by the Pu^{242} as compared to that from the much more abundant Pu^{239} activity.

Correlating Am^{243} With Cm^{244}

These two, chemically similar rads were produced in SRS reactors during high flux irradiation of plutonium targets enriched in Pu^{242} . The Cm-II campaign produced the bulk of these that have been processed at SRS. These targets underwent chemical processing in H-Canyon and then in the High Level Caves in SRTC to separate and purify approximately 3.5 kg of Cm^{244} oxide as well as some mixed oxides of Am^{243} and Cm^{244} . The date of discharge from K Reactor for the Cm-II charge was May 10, 1967.⁸ This date is used for $t = 0$ in the standard decay equation to develop the data shown in Table 3.

Other assumptions used were:

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1. There was no separation of Am^{243} from Cm^{244} during processing or during environmental residence. Such an assumption is valid since most waste produced from processing of Cm-II material was produced prior to separation of the two elements. Separation of Am^{243} from Cm^{244} did occur in the final stages of SRTC processing just prior to preparing the oxides.⁹
2. The mass ratio at discharge was 2 g Cm^{244} /0.9 g Am^{243} . This value is actually documented¹⁰ and not an assumption.
3. The mobility of these two rads in soils and groundwater is equivalent. No chemical separation occurs. These assumptions are also valid because both elements exist primarily as trivalent ions in solution and have quite similar chemical properties. Americium can exist in the +6 state in solution as the americyl ion, AmO_2^{+2} , but it is such a strong oxidizing agent that it is very easily reduced to the trivalent oxidation state. In basic media, americium can also exist as the pentavalent ion, AmO_2^+ , but only in strong oxidizing conditions (using ozone or persulfate oxidants) that are unlike any normally found in the environment.

The activity ratio in Table 3 is plotted versus time in Figure 3. This dramatically shows the very low activity of the Am^{243} as compared with the Cm^{244} now and in the few decades to come. This case is similar to the $\text{Pu}^{242}/\text{Pu}^{239}$ activity comparison in the preceding section.

CONCLUSIONS

- For the next decade, the activity of any Co^{60} found in groundwater samples can be used to estimate the activity of any Ni^{63} present. The current activity ratio for $\text{Ni}^{63}/\text{Co}^{60}$ is approximately 1.
- For at least the next thirty years, the activity of any Pu^{239} found in groundwater samples can be used to estimate the activity of any Pu^{241} present. The current activity ratio for $\text{Pu}^{241}/\text{Pu}^{239}$ is approximately 4.

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- For the next 1000 years at least, the activity of any Pu²³⁹ found in groundwater samples can be used to estimate the activity of any Pu²⁴² present. The current activity ratio for Pu²⁴²/Pu²³⁹ is approximately 2E-05.
- For the next 80 years, the activity of any Cm²⁴⁴ found in groundwater samples can be used to estimate the activity of any Am²⁴³ present. The current activity ratio for Am²⁴³/Cm²⁴⁴ is approximately 3E-03.

This approach to evaluating rad data can be used to ascertain which rads are important in certain relationships. For example, if Sr⁹⁰ is being analyzed for, is the Sr⁸⁹ analysis truly necessary since it has a much shorter half life? Cost savings can be realized by eliminating analysis of the unimportant rads or by being able to estimate the activities of those of concern. I plan to investigate these potentially useful correlations and report on them at a future date.

References

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2. H. B. Peacock, Jr. to H. P. Holcomb, Private Communication, Apr. 7, 1993.
3. Bob Brown, Carpenter Steel to H. P. Holcomb, Private Communication, Apr. 13, 1993.
4. N. P. Baumann to H. P. Holcomb, "Ni⁶³/Co⁶⁰ (U)", May 5, 1993.
5. Wendell M. Latimer, Oxidation Potentials, 2nd Ed. p. 210, Prentice-Hall, Inc. New Jersey (1952).
6. Ibid., pp. 200, 201, 212, 213.
7. P. L. Fisk to H. P. Holcomb, SET-SEP-930032, "FB-Line Safety Analysis Report Source Term Isotopics (U)", Apr. 6, 1993.

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8. J. L. Jarriel to Distribution, OPS-PSD-910032, "Reactor Operating History (U)", Table 4, March 28, 1991.

9. I. D. Eubanks and G. A. Burney, DP-1009, "Curium Process Development. I. General Process Description" (U), 1966.

10. W. E. Prout, H. E. Henry, H. P. Holcomb, and W. J. Jenkins, DP-1302, "Curium Process Development. II-2. Recovery of Curium and Americium by Solvent Extraction with Tertiary Amines" (U), Table 1, December, 1972.

Table 1

CALCULATION OF Ni-63 ACTIVITY FROM Co-60 ACTIVITY				
Time (years) Since Reactor Irradiation Ceased	Calendar Year (January)	Equivalent Ni-63 Activity Present	Equivalent Co-60 Activity Present	Activity Ratio, Ni-63/Co-60
0	1971	.05723*	1	0.05723
5	1976	0.05528094	0.51814777	0.106689535
10	1981	0.05339827	0.26847711	0.198893183
15	1986	0.05157971	0.13911081	0.370781429
20	1991	0.04982308	0.07207996	0.691219608
25	1996	0.04812628	0.03734807	1.288588127
30	2001	0.04648726	0.01935182	2.402216807
35	2006	0.04490407	0.0100271	4.47827003
40	2011	0.04337479	0.00519552	8.348498106
45	2016	0.0418976	0.00269205	15.56346986
50	2021	0.04047071	0.00139488	29.01379277
100	2071	0.02861923	1.9457E-06	14709.07165
250	2221	0.01012067	5.2806E-15	1.91658E+12
500	2471	0.00178976	2.7885E-29	6.41847E+25
750	2721	0.0003165	1.4725E-43	2.14949E+39
1000	2971	5.5971E-05	7.7755E-58	7.19845E+52
*Activity ratio calculated by N. P. Baumann, SRTC.				
Data bases contained in memorandum N. P. Baumann to H. P. Holcomb, "Ni-63/Co-60", May 5, 1993.				
Fractions remaining calculated from standard decay equation.				
Assume: Migratory Ni-63 and Co-60 were present in a SRS reactor that shutdown (irradiation ceased) on Jan. 1, 1971. This is approximately mid-way during the production mode operating period for SRS Reactors (1954-1988).				
Assume: No separation of the Ni-63 and Co-60 occurred during processing or during environmental residence because of their similar chemistry.				
Assume: Half-lives are: Ni-63 = 100 years; Co-60 = 5.27 years.				
Ref: Edgardo Browne and Richard B. Firestone, "Table of Radioactive Isotopes", p. 63-1 and p. 60-1, John Wiley and Sons, New York (1986)				

Figure 1

Ni-63/Co-60 Activity vs Time

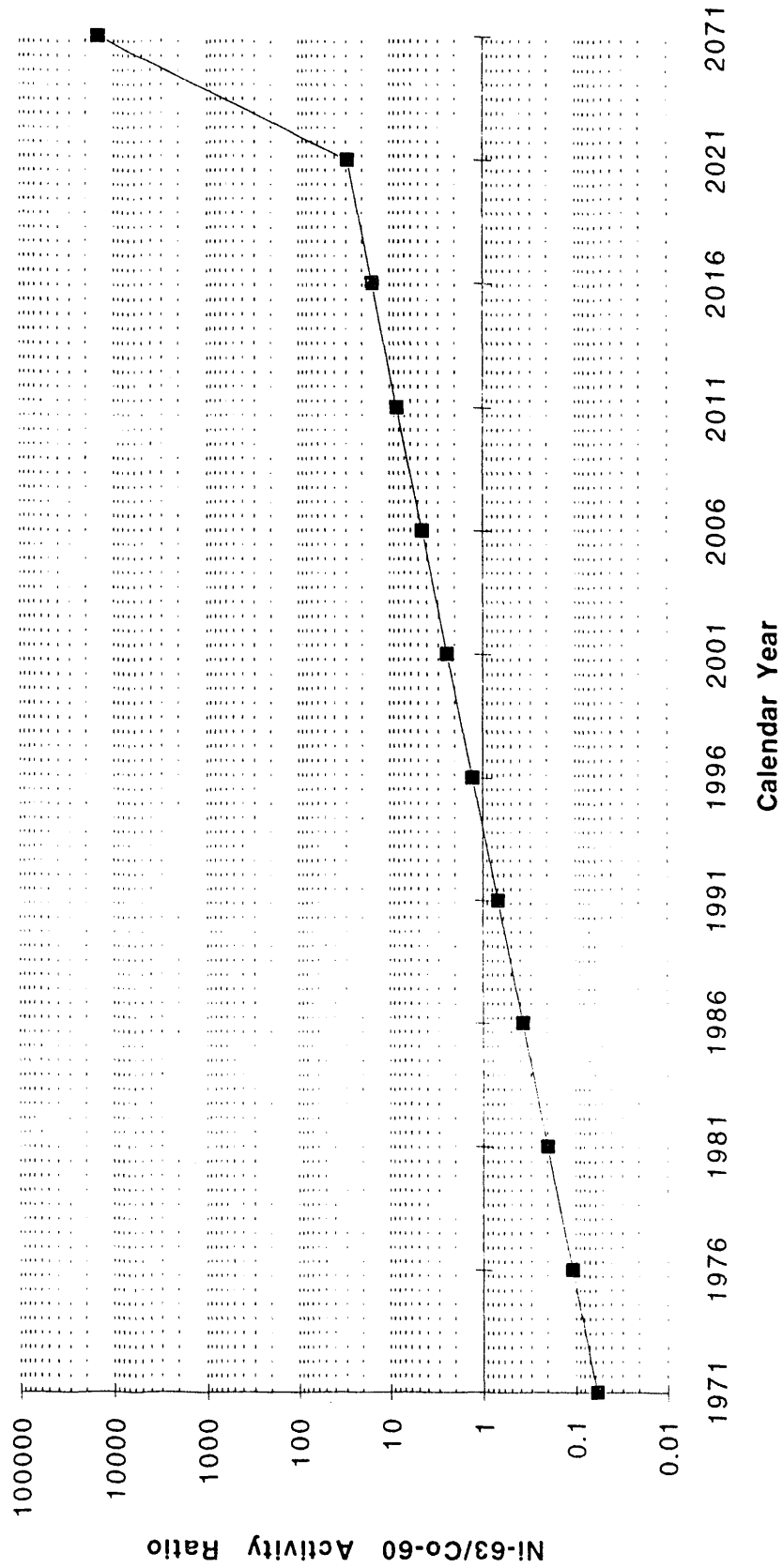


Table 2
 CALCULATION OF Pu-241 AND Pu-242 ACTIVITIES FROM Pu-239 ACTIVITY

Assume: 10000 g Weapons Grade Plutonium as source term at reactor shutdown, 01/01/71											
Time (years) Since Reactor Irradiation Ceased	Calendar Year (January)	Pu-239, g (Remainder due to Pu-238 and Pu-240)	Pu-241, g	Pu-242, g	Pu-239 Activity, (d/m/grams in Col. 3)	Pu-241 Activity, (d/m/grams in Col. 4)	Pu-242 Activity, (d/m/grams in Col. 5)	Pu-239 Activity (Col. 7/Col. 6)	Pu-241 Activity (Col. 8/Col. 6)	Pu-242 Activity (Col. 8/Col. 6)	Activity/ (Col. 8/Col. 6)
0	1971	9341	70	3	1.2853E-15	1.6002E+16	2.613E+10	12.44980245			2.03295E-05
5	1976	9339.65764	55.029555	2.99997174	1.2851E+15	1.258E+16	2.613E+10	9.788650801			2.03323E-05
10	1981	9338.31548	43.2607417	2.99994348	1.285E+15	9.8994E+15	2.613E+10	7.696321682			2.0335E-05
15	1986	9336.97351	34.0088481	2.99991521	1.2848E+15	7.7744E+15	2.6129E+10	6.051228984			2.03377E-05
20	1991	9335.63173	26.7355968	2.99988695	1.2846E+15	6.1118E+15	2.6129E+10	4.757775691			2.03405E-05
25	1996	9334.29015	21.0178284	2.99985869	1.2844E+15	4.8047E+15	2.6129E+10	3.740798702			2.03432E-05
30	2001	9332.94875	16.5228821	2.99983043	1.2842E+15	3.7771E+15	2.6129E+10	2.941201065			2.03459E-05
35	2006	9331.60755	12.9892407	2.99980217	1.284E+15	2.9693E+15	2.6128E+10	2.312517832			2.03487E-05
40	2011	9330.26655	10.2113162	2.99977391	1.2838E+15	2.3343E+15	2.6128E+10	1.818215962			2.03514E-05
45	2016	9328.92573	8.02748837	2.99974565	1.2837E+15	1.8351E+15	2.6128E+10	1.429571369			2.03541E-05
50	2021	9327.58511	6.31070161	2.99971739	1.2835E+15	1.4426E+15	2.6128E+10	1.123999757			2.03569E-05
100	2071	9314.18949	0.56892793	2.99943481	1.2816E+15	1.3006E+14	2.6125E+10	0.10147755			2.03842E-05
250	2221	9274.11794	0.00041687	2.99858722	1.2761E+15	9.5295E+10	2.6118E+10	7.4676E-05			2.04665E-05
500	2471	9207.71475	2.4825E-09	2.9971751	1.267E+15	567505.826	2.6105E+10	4.4792E-10			2.06044E-05
750	2721	9141.78702	1.4784E-14	2.99576365	1.2579E+15	3.37962375	2.6093E+10	2.6867E-15			2.07432E-05
1000	2971	9076.33133	8.8042E-20	2.99435286	1.2489E+15	2.0126E-05	2.6081E+10	1.61153E-20			2.0883E-05

Assume: For the source term, weapons-grade plutonium with the following isotopic weight percents at reactor shutdown: Pu-238 - 0.007, Pu-239 - 93.41, Pu-240 - 5.86, Pu-241 - 0.70, and Pu-242 - 0.03. Such an assumption is valid because most of the plutonium produced at SRS has been weapons grade with nominally 6% or less Pu-240.

Ref: SRT-SEP-93-0032, "FB-Line Safety Analysis Report Source Term Isotopics (U)".
 P. L. Fisk, April 6, 1993

Assume: Migratory plutonium originated in reactor in which irradiation ceased on Jan. 1, 1971. This is approximately mid-way during the production mode operating period for SRS Reactors (1954-1988).

Assume: No isotopic separation occurred during processing or during environmental residence. This assumption is quite sound because isotopic separation of plutonium in the environment or during SRS processing is unknown.

Assume: Half-lives are: Pu-239 - 2.411E04 years, Pu-241 - 14.4 y, and Pu-242 - 3.763E05 years which result in following specific activities (d/m/g): Pu-239 - 1.376E11; Pu-241 - 2.286E14; and Pu-242 - 8.710E09

Ref: Edgardo Browne and Richard B. Firestone, "Table of Radioactive Isotopes", p. 239-2, p. 241-1, and p. 242-2, John Wiley and Sons, New York (1986)

Figure 2

Pu-241/Pu-239 and Pu-242/Pu-239 Activity Relationships vs Time

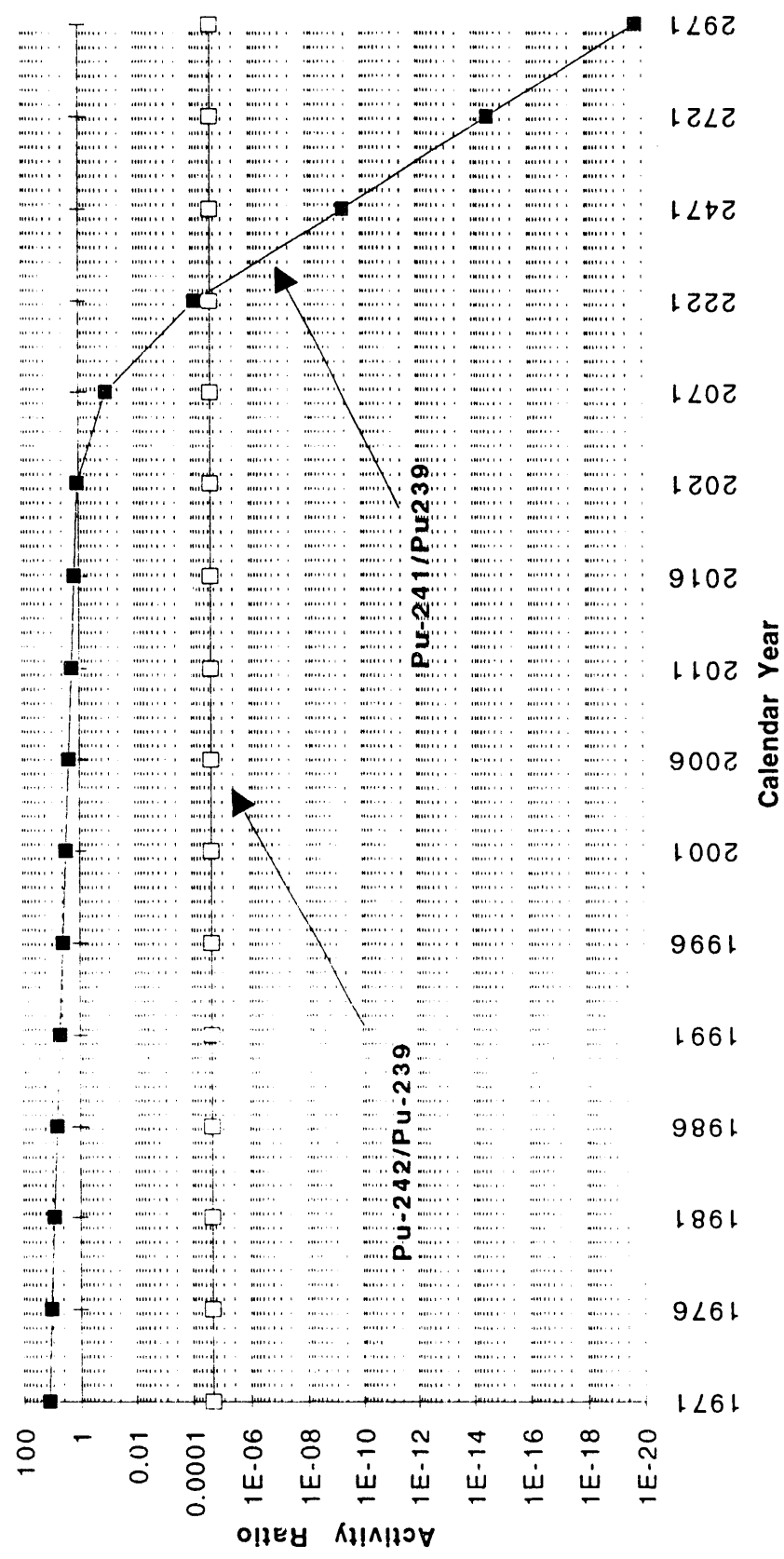
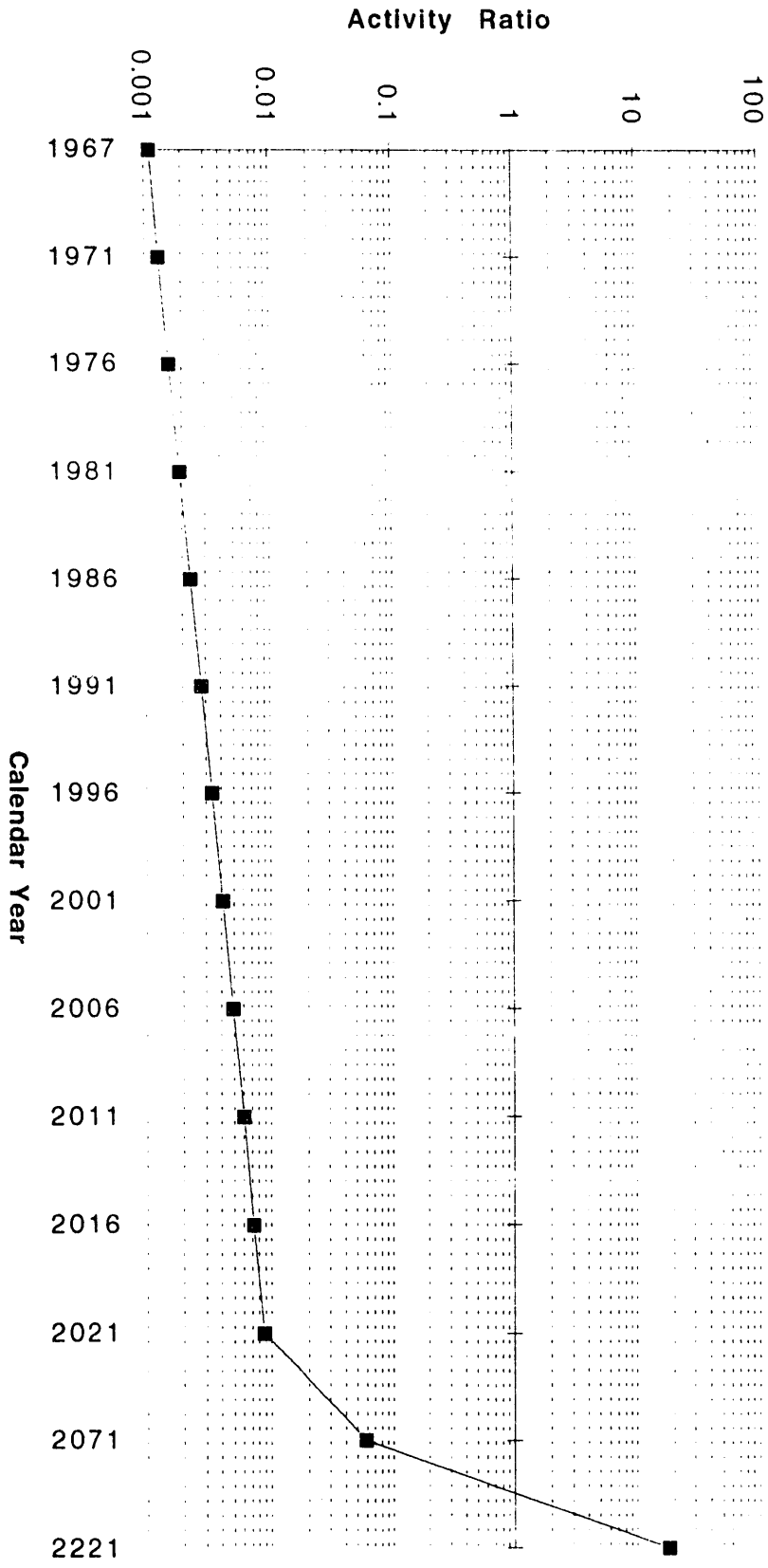


Table 3

CALCULATION OF Am-243 ACTIVITY FROM Cm-244 ACTIVITY						
Assume: 100 g Cm-244/45 g Am-243 Mass Ratio at Cm-II Discharge, May 10, 1967 = (t = 0)						
Time, years Since Reactor Discharge	Calendar Year	Cm-244, g	Am-243, g	Equivalent Cm-244 d/m/grams in Col. 3	Equivalent Am-243 d/m/grams in Col. 4	Am-243 Activity/ Cm-244 Activity (Col. 6/Col. 5)
0	1967	100	45	1.80E+16	1.99E+13	0.0011075
4	1971	85.8074372	44.9831007	1.54E+16	1.99E+13	0.001290196
9	1976	70.8648864	44.9619856	1.28E+16	1.99E+13	0.001561513
14	1981	58.524439	44.9408804	1.05E+16	1.99E+13	0.001889886
19	1986	48.3329634	44.919785	8.70E+15	1.99E+13	0.002287312
24	1991	39.9162366	44.8986996	7.18E+15	1.99E+13	0.002768314
29	1996	32.9652028	44.8776241	5.93E+15	1.99E+13	0.003350467
34	2001	27.2246256	44.8565585	4.90E+15	1.99E+13	0.004055041
39	2006	22.4837154	44.8355027	4.05E+15	1.99E+13	0.004907781
44	2011	18.5683898	44.8144569	3.34E+15	1.99E+13	0.005939845
49	2016	15.3348809	44.7934209	2.76E+15	1.98E+13	0.007188943
54	2021	12.664457	44.7723948	2.28E+15	1.98E+13	0.008700716
104	2071	1.86916747	44.5626758	3.36E+14	1.97E+13	0.058675158
254	2221	0.00600943	43.9393945	1.08E+12	1.95E+13	17.99501243
504	2471	4.2086E-07	42.9199045	7.58E+07	1.90E+13	250985.495
754	2721	2.9475E-11	41.9240689	5.31E+03	1.86E+13	3500621017
1004	2971	2.0642E-15	40.9513388	3.72E-01	1.81E+13	4.88249E+13
Assume: Bulk of source term for Cm-244 and Am-243 at SRS was produced in the Cm-II Campaign which generated some 3.5 kg of Cm-244 which was shipped to ORNL for HIFR Target material and for Isotope Sales.						
Assume: Date of discharge for Cm-II from K Reactor was May 10, 1967. Ref: OPS-PSD-91-0032, "Reactor Operating History (U)", Table 4, J. L. Jarriel, 3/28/91						
Assume: For the source term, no separation of Curium from Americium occurred during SRS processing. Such an assumption is valid since most waste produced resulted from processing of Cm-II material prior to separation of the two elements. Only in the final stages of processing in the SRTC High Level Caves did their separation take place. And even there, not all the Am was separated from the Cm since some mixed oxides were produced. Ref.: DP-1009, "Curium Process Development. I. General Process Description," I. D. Eubanks and G. A. Burney, 1966.						
Assume: Cm-244/Am-243 at discharge = 2g/0.9g Ref: DP-1302, "Curium Process Development II-2. Recovery of Curium and Americium by Solvent Extraction with Tertiary Amines (U)", Table I, W. E. Prout, H. E. Henry, H. P. Holcomb and W. J. Jenkins, December 1972.						
Assume: Half Life for Cm-244 = 18.11 y and for Am-243 = 7380 y and specific activities are 1.80E14 d/m/g for Cm-244 and 4.43E11 d/m/g for Am-243 based on these half lives. Ref: Edgardo Browne and Richard B. Firestone, "Table of Radioactive Isotopes", p. 244-2 and p. 243-1, John Wiley and Sons, New York (1986)						
Assume: Mobility of Cm and Am in soils is equivalent, i. e., no chemical separation occurs. Both elements exist primarily as trivalent ions in solution and have quite similar chemical properties making such an assumption valid. Higher oxidation states of Am are known, but they can only exist in highly oxidizing conditions.						

Figure 3



Am-243/Cm-244 Activity Relationships vs Time

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