Health Physics Pergamon Press 1966. Vol 12, pp. 1545–1555. Printed in Northern Ireland

A COMPARISON OF URANIUM CASES SHOWING LONG CHEST BURDEN RETENTIONS

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(*Received* 10 *December* 1965; *in revised form* 9 *March* 1966)

Abstract—Routine *in vivo* gamma spectrum measurements have been made to estimate internal exposure to uranium for approximately 2500 persons at the United States Atomic Energy Commission's Y-12 Plant. With only five exceptions, all cases exhibited normal elimination patterns. The five exceptions showed extended biological half-lives of from 380 to 1470 days. The detection of these cases was possible because of the use of the direct-measuring technique of *in vivo* spectrometry. *In vivo*, urine and fecal data on these persons have been obtained and evaluated. The uranium urine excretion rate was found to decrease more rapidly than the chest burden. The urine and fecal elimination rate on these employees after their removal from uranium exposure was determined to be about equal.

EVALUATION OF THE CASES

Introduction

Background. For years, health physicists in the uranium industry depended almost exclusively on urine analysis to estimate internal exposure to uranium. More recently, *in vivo* gamma spectrometry utilizing the 186-keV gamma radiation associated with the uranium-235 isotope has been used to evaluate uranium chest burdens. Cases involving an elevated chest burden detected by such a technique have been reported by FISH,[1] SAXBY *et al.*[2,3] and SCOTT and WEST.[4,5]

In the Y-12 Plant, this technique has been used routinely for the last 4 years in estimating the internal exposure to uranium of some 2500 employees.

Scope and purpose. Of the cases showing detectable chest burdens during this period, five have indicated burdens which exhibit an unexpectedly slow decrease after the persons were assigned to nonuranium activities.

The purposes of this report are to: (1) present the *in vivo*, urine and fecal monitoring results on these persons; (2) compare these data to that from similar cases previously reported; and (3) describe briefly the continuing efforts that are being made to delineate the reasons for these unusual retention patterns.

Data

In vivo data. Tables 1 through 5 give the individual *in vivo* datum for the five cases under consideration. Urine and fecal data, averaged for monthly periods, are shown for comparison. Examination of these data will show that the variability of the excretion data, especially the fecal, is relatively greater than that for *in vivo* data.

The data in these tables were used to derive the best fitting exponential curves by computer programing. Computer computations and visual checks of the plotted data indicated that this single exponential equation fit was as good or better than that of a power function or multiple exponential function for these data. Consequently, for ease and clarity of presentation all graphs in this report present the data in the form of best fit curves derived in this fashion.

Figure 1 shows the curves for the *in vivo* data. The slopes give a visual index of the half-life which is shown in parentheses after each case designation. The shortest half-life is 380 days; the longest 1470 days. The abscissa shows how long these employees have been monitored since their first elevated *in vivo* measurement. Subsequent measurements were made on a monthly basis.

Figure 2 compares the Y-12 cases with the

Table 1. *Case* Y-I

In vivo			Urine		Feces	
Date	pc	Month	Number of samples	Average pc/day	Number of samples	Average pc/day
1-08-62	37,500					
1-29-62	34,700					
2-05-62	41,000	2-62	4	41.0		
2-13-62	41,800					
2-20-62	40,300*					
3-02-62	43,900	3-62	3	54.4		
3-23-62	44,900					
4-25-62	35,900	4-62	5	40.5		
		5-62	3	27.4		
		6-62	3	13.5		
		7-62	4	12.2		
		8-62	4	18.9		
9-24-62	33,900	9-62	3	19.4		
10-10-62	28,200	10-62	3	9.9		
11-06-62	30,900	11-62	6	14.4	5	9.1
12-01-62	32,600					
1-08-63	34,200	1-63	1	11.3		
2-12-63	31,800	2-63	3	5.8		
3-28-63	27,900	3-62	3	15.3	1	5.4
		4-63	6	14.4	1	4.1
5-07-63	36,000	5-63	2	13.9	2	7.2
		7-63	2	19.8		
		8-63	1	6.7		
10-09-63	17,200					
12-04-63	17,500	12-63	4	11.2		
1-06-64	17,300	1-64	4	7.2		
2-10-64	21,700	2-64	2	9.0		
3-25-64	11,800	3-64	5	11.3		
		4-64	3	2.2		
5-20-64	14,400	5-64	3	7.6		
6-19-64	16,500	6-64	3	8.5		
6-30-64	13,100					
		7-64	4	5.4		
8-21-64	12,800	8-64	3	13.5		
		9-64	2	2.2		
10-07-64	10,700	10-64	2	6.7		
11-18-64	17,500	11-64	2	9.4		
		12-64	2	13.0		

* Designates last count before removal of employee from uranium handling areas.

Table 2. *Case* Y-2

In vivo Date	*In vivo* pc	Month	Urine: Number of samples	Urine: Average pc/day	Feces: Number of samples	Feces: Average pc/day
		8-62	4	45.0		
		9-62	3	39.1		
10-08-62	39,500*	10-62	5	36.4		
10-19-62	42,300					
11-05-62	44,300	11-62	3	18.9		
11-12-62	46,500					
11-29-62	38,800					
12-09-62	34,700	12-62	4	38.7		
12-29-62	44,100					
1-15-63	38,800	1-63	3	22.9	5	44.5
2-12-63	38,700	2-63	3	25.6	2	37.1
3-12-63	36,000	3-63	3	20.2	4	18.2
4-23-63	33,900	4-63	4	11.7	2	14.6
		5-63	2	3.1		
		6-63	2	22.9		
		7-63	2	5.4		
8-14-63	30,800	8-63	2	11.7		
9-27-63	33,300	9-63	2	10.2	3	7.4
		10-63	1	7.2	1	10.4
11-20-63	33,100	11-63	1	10.3	1	14.9
12-20-63	30,200					
2-12-64	26,500	2-64	3	9.4	2	5.9
3-19-64	26,600	3-64	3	7.2	3	11.8
		4-64	2	11.7	3	4.5
5-06-64	24,100	5-64	2	8.5	1	7.7
6-04-64	27,500	6-64	3	8.5	1	4.5
7-14-64	28,600	7-64	1	17.5	1	8.1
8-27-64	26,400	8-64			1	9.0
9-28-64	27,600	9-64	1	9.0	1	11.3
		10-64			1	18.5
11-23-64	21,600					
12-30-64	21,100	12-64	2	7.2	1	5.9
1-25-65	23,900					

* Designates last count before removal of employee from uranium handling areas.

Table 3. *Case* Y-3

In vivo			Urine		Feces	
Date	pc	Month	Number of samples	Average pc/day	Number of samples	Average pc/day
1-11-63	22,200	1-63	4	16.2		
		2-63	3	26.1		
3-01-63	21,400	3-63	4	22.9		
4-02-63	21,100*	4-63	4	51.7		
5-09-63	22,300	5-63	3	22.0		
5-24-63	13,100					
		6-63	3	18.9		
7-25-63	13,700	7-63	4	14.8		
		8-63	2	15.3		
9-27-63	7200	9-63	5	8.1		
		10-63	2	12.6		
11-18-63	16,500	11-63	1	10.8		
12-20-63	12,100	12-63	2	7.6	3	22.2
1-28-64	9000	1-64	3	5.4	4	9.8
		2-64	4	5.4	2	17.6
		3-64	5	10.3	3	12.5
4-03-64	11,700	4-64	3	6.3	2	17.8
5-19-64	14,800	5-64	5	5.4	2	9.2
6-09-64	11,300					
6-19-64	16,100					
7-21-64	14,300					
8-21-64	5900	8-64	9	8.5	2	6.5
9-21-64	7800	9-64			2	8.3
10-21-64	8400	10-64	4	4.0	2	12.6
11-27-64	4800	11-64			1	13.5
12-14-64	4100	12-64	4	12.1	2	6.4
12-28-64	10,800					
		1-65	4	5.0	4	10.2
2-08-65	3500					

* Designates last count before removal of employee from uranium handling areas.

longest and shortest half-life to a reference line which starts at the level which would give the lung a dose of 15 rem/yr and drops with the half-life of 120 days assumed for insoluble uranium by both the ICRP[6] and NCRP[7] in calculating uranium air limits with the lung as the critical organ. The cases reported by Fish (Case 1) and Saxby (Case UR-2) are also shown. Cases 1 and UR-2 show one or more faster components. However, their longest component with half-lives of 390 and 300 days, respectively, are comparable in half-life with Case Y-3 but are much shorter in half-life than the other "Y" cases.

Urinalyses. These five employees were participating in a routine urinalysis program at the time it was revealed that their *in vivo* monitoring results were elevated. They have continued on such a program but have shown no urine results of the magnitude usually considered indicative of the likelihood of a significant lung burden.

Figure 3 shows the plots of the monthly averages of urine samples given by these persons. These exponential functions show half-lives of from 300 to 460 days, indicating a faster decline in the excretion rate than in the chest burden.

Figure 4 compares two representative functions from Fig. 3 with the urinalyses excretion curve for a lung burden amount based on a 120-day half-life and two-thirds of the uranium being eliminated from the lung being excreted in the urine. Also compared is Case 1, which shows the highest excretion rate of all cases under

Table 4. Case Y-4

In vivo			Urine		Feces	
Date	pc	Month	Number of samples	Average pc/day	Number of samples	Average pc/day
2-25-63	26,200*	2-63	4	10.8		
		3-63	3	13.0		
4-03-63	26,500	4-63	4	13.5	2	9.9
4-23-63	20,800					
5-24-63	18,600	5-63	4	9.4	4	7.8
		6-63	4	8.5	4	5.6
7-24-63	22,400	7-63	3	10.8	3	9.0
		8-63			4	5.5
		9-63	5	6.3		
10-02-63	18.300	10-63	1	12.6	1	4.1
11-20-63	23,500	11-63	3	9.4	2	7.0
12-27-63	25.000	12-63	4	6.7		
1-29-64	16.900	1-64	4	5.8		
2-27-64	7300	2-64			2	8.3
3-30-64	23,800	3-64	11	3.4		
		4-64			1	4.9
5-11-64	20,700	5-64	3	1.8		
6-09-64	22,400	6-64	4	6.7		
7-21-64	18,600	7-64	2	4.9		
8-21-64	20,000	8-64	5	1.8	1	6.7
		9-64	4	3.1	1	6.7
10-21-64	20,400	10-64				
11-20-64	13,300	11-64	5	6.7		
12-09-64	17,500	12-64	3	6.7		
1-08-65	15,200					
2-16-65	18,200					

* Designates last count before removal of employee from uranium handling areas.

Table 5. Case Y-5

In vivo			Urine		Feces	
Date	pc	Month	Number of samples	Average pc/day	Number of samples	Average pc/day
11-18-63	23,000					
12-03-63	25,200*	12-63	3	9.0		
1-06-64	18,200	1-64	2	6.3		
1-10-64	22,600					
2-10-64	20,100	2-64	4	10.3	4	26.4
3-10-64	25,300	3-64			3	7.5
6-02-64	27,400	6-64	3	2.1		
7-02-64	20,200	7-64	3	3.1		
8-03-64	16,700	8-64	4	2.2	3	27.0
9-03-64	17,500	9-64	3	4.5	3	15.9
10-01-64	21,200	10-64	9	3.7		
11-25-64	15,000					
12-09-64	16,700					
1-08-65	23,400	1-65	2	6.4	1	7.2
2-15-65	18,700					

* Designates last count before removal of employee from uranium handling areas.

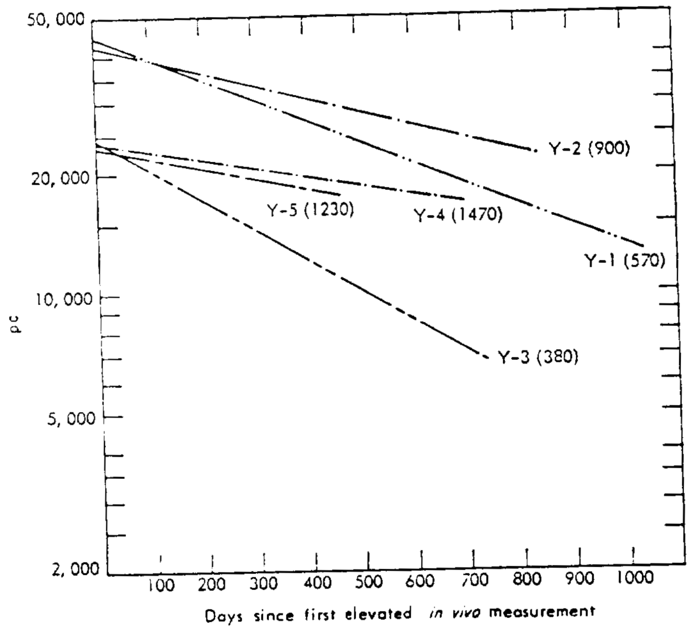

FIG. 1. *In vivo* measurement of chest burden.

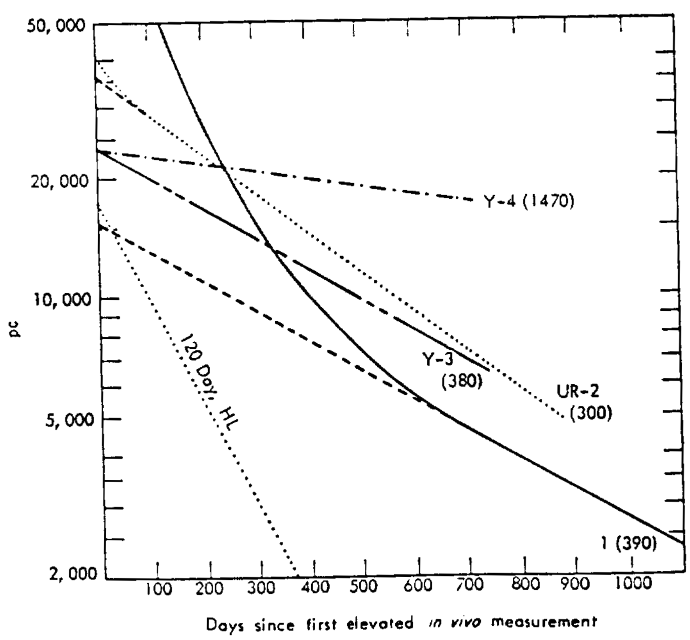

FIG. 2. *In vivo* measurement of chest burden (comparison).

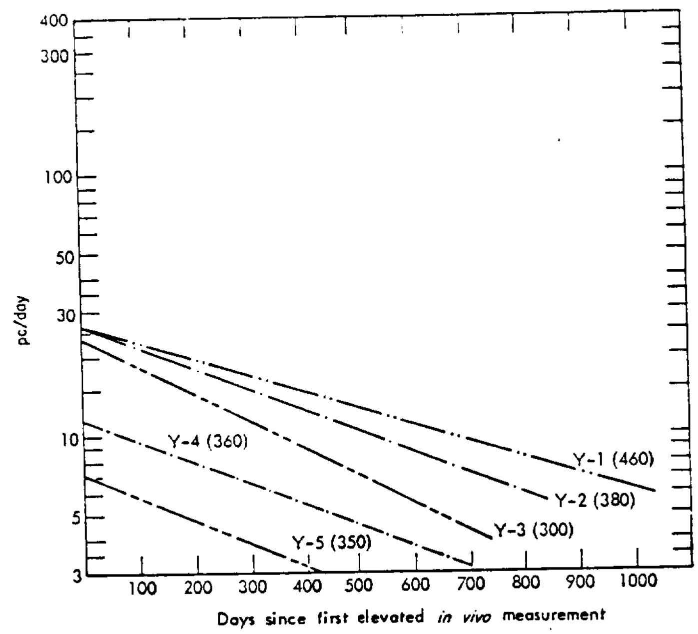

FIG. 3. Urinalyses.

consideration and two faster exponential components than the longest component of 240 days. The urine excretion of UR-2 falls within the range of the other cases in magnitude and half-life. Case UR-2[2] showed an earlier 12-day half-life component which is not seen in this figure because its effect was spent prior to the first body count which came some 67 days after exposure.

Comparison of in vivo and urine data. Estimation of the chest burden from urinalyses results requires the assumption of some ratio between burden and excretion rate. Table 6 presents a tabular comparison of the ratios of burden to urine excretion rate obtained in these cases for the periods from 1 to 1000 days as data are available. Note that all cases increase in ratio between the initial and final determination, and that there is considerable variability between individuals giving ratios which range from 270 to 5700.

Fecal analyses. Uranium analyses have been made on fecal samples from these individuals for a substantial portion of the time since their removal from uranium exposure.

Figure 5 shows the function for the monthly averages of the fecal data and illustrates the periods of time covered. These fecal excretion curves vary in half-life from 260 to 1930 days. Figure 6 compares two cases from Fig. 5 with Cases UR-2 and 1. Case UR-2 shows a higher initial level of 200 pc and a two-component half-life curve with half-lives of 30 and 280 days. There is too little fecal data in Case 1 for meaningful curve comparison, but the level obtained during the short period of sampling is shown.

Comparison of excretion and in vivo data. The fecal-to-urine ratios for all cases are given in Table 7. In every case except Case 1, this ratio shows feces elimination to be a significant means of clearance, proving that weight must be given this mode of elimination when considering excretion limits or retention parameters. As previously shown in Fig. 6, the fecal sampling in Case 1 covers only a very short period of time (i.e. five samples over a 3-day period). The ratios of the total excretion rates to the *in vivo* measurement, for the periods for which fecal data are available, are reported in Table 8. These ratios consistently increase with time since exposure, showing further that excretion is decreasing more rapidly than chest burden. However, this situation could change with time on

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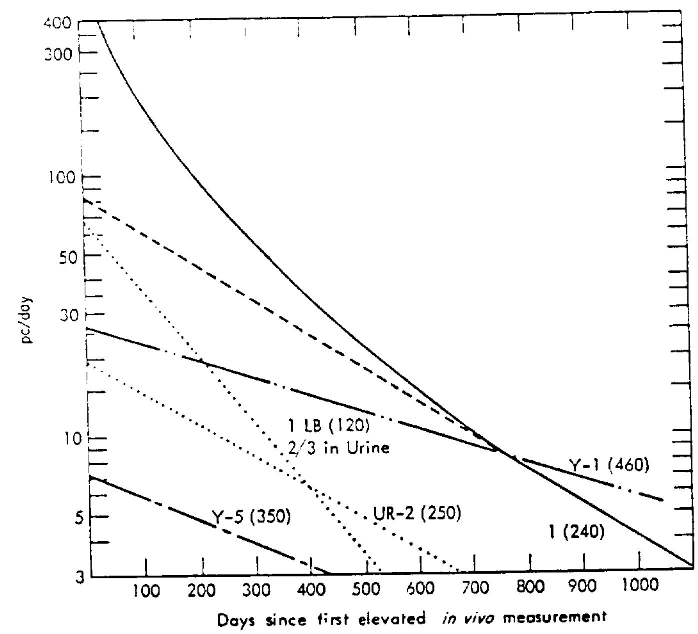

FIG. 4. Urinalyses (comparison).

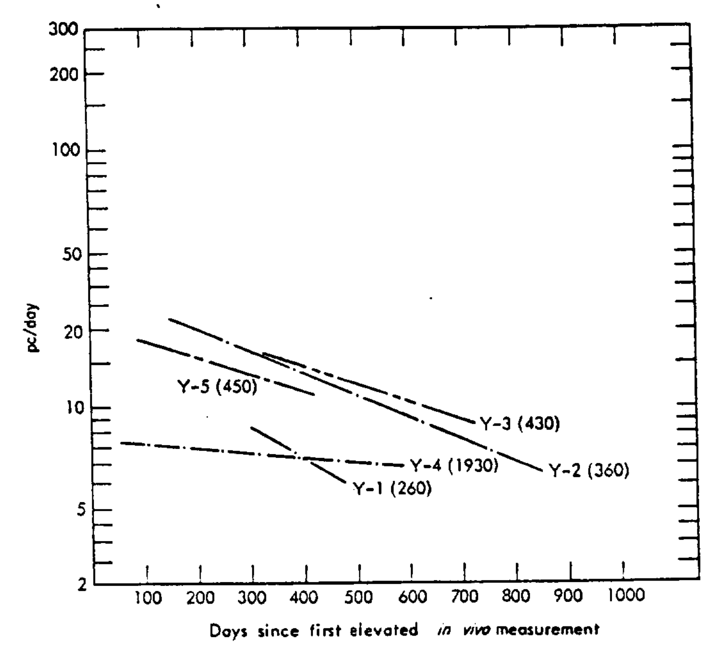

FIG. 5. Fecal analyses.

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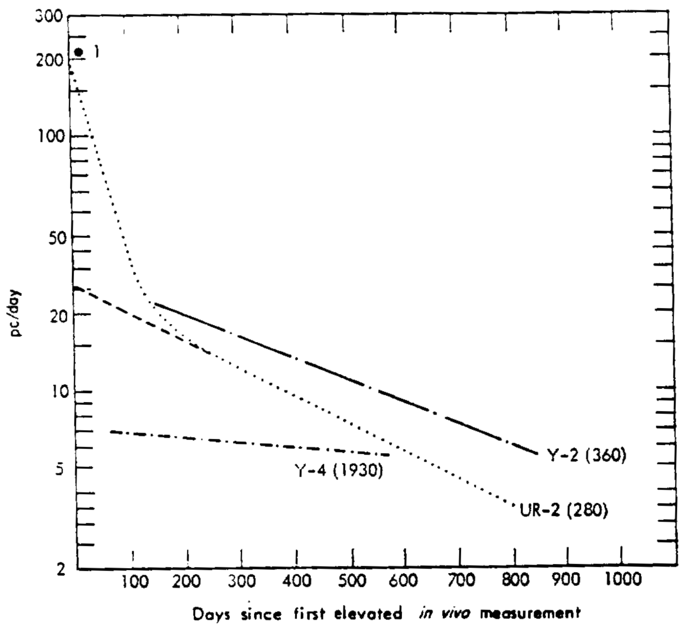

Fig. 6. Fecal analyses (comparison).

Table 6. *Ratio of* in vivo *measurement to urinalyses excretion rate*

	Case						
Ratio*	1	Y-3	Y-1	UR-2	Y-2	Y-4	Y-5
Initial	270	1100	1700	2200	1600	2100	3200
Final	610	1600	2200	3100	3800	4900	5700

* Chest burden (pc)/urine excretion rate (pc/day).

Table 7. *Ratio of fecal excretion to urine excretion (integral of functions over period covered by fecal analyses)*

	Case						
	1	Y-1	Y-2	Y-4	UR-2	Y-3	Y-5
Ratio*	0.1	0.5	0.9	1.0	1.1	1.6	3.3

* Fecal excretion (pc)/urine excretion (pc).

Table 8. *Ratio of* in vivo *measurements to total excretion rate*

	Case						
Ratio*	1	Y-3	UR-2	Y-5	Y-1	Y-2	Y-4
Initial	NA†	520	200	920	1200	900	1300
Final	NA†	540	1200	1300	1400	1800	1800

* Chest burden (pc)/urine plus fecal excretion rate (pc/day).
† Not available.

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Table 9. Ratio of drop in chest measurements to total excreted (for periods of time covered by fecal analyses)

	Case						
	Y-4	Y-5	1	Y-3	Y-2	Y-1	UR-2
Ratio*	0.7	0.7	0.9	0.9	1.1	1.6	2.6

* Drop in chest measurements (pc)/sum of integral of urine and fecal functions (pc).

Cases Y-3 and Y-4 since their fecal-excretion function shows a longer half life than does their chest-retention function.

A material balance was calculated on all cases for the periods covered by fecal analysis. The ratio of the amount of drop shown by *in vivo* to total excreted is shown in Table 9. These ratios are well within the limits of error of these measurements with the exception of Cases Y-1 and UR-2. The imbalance in Case Y-1 is believed to hinge around the fecal data. Note from Table 1 that the fecal data in this case are sparce and cover only a short period of time. Table 7 shows that Y-1 had the lowest fecal-to-urine excretion rate ratio of the Y cases. These facts suggest that fecal data on this case underestimate the actual fecal excretion.

The imbalance in the case of UR-2 is believed to be the result of *in vivo* measurements which are low. In all the Y cases, unexpected and unexplained drops in body count (greater than could be accounted for by statistical variation) have been experienced. These low measurements have been followed by measurements which fit the general pattern of the rest of the *in vivo* data. Examples are: (1) the drop of 9000 pc between 3-23-62 and 4-25-62 in Case Y-1; (2) the drop of almost 18,000 pc between 12-27-63 and 2-27-64 in Case Y-4; (3) the drop of 11,800 pc between 11-12-62 and 12-9-62 in Case Y-3; and (4) the drop of 7000 pc between 12-3-63 and 1-6-64 in Case Y-5. It is predicted that if subsequent *in vivo* measurements are made on UR-2 that they will indicate a higher level and consequently there will be a better material balance.

INVESTIGATION OF THE CASES

A detailed compilation of work and exposure histories on the five Y cases and comparison with other employees doing similar work has failed to reveal either the specific source or nature of the exposure. Comprehensive physical examinations of the people involved revealed no abnormalities which would help to explain the long biological half-lives exhibited, or any detectable physical harm from the exposures incurred.[8] The physical and chemical properties of likely exposure material are presently being studied in detail. Such a study may lead to an investigation by animal experimentation to help define which exposure materials give such retention patterns.

SUMMARY AND CONCLUSIONS

Summary

Five persons out of some 2500 monitored by *in vivo* spectrometry for uranium in the Y-12 Plant have shown chest burdens with biological half-lives ranging from 380 to 1470 days. Monitoring data from these persons show ratios of chest burden to urine excretion rate of from 270 to 5700. Comparison with cases previously reported reveals that the Y cases generally show retention functions with longer half-lives. Excretion functions in the Y cases did not exhibit the initial shorter half-live components found in Cases 1 and UR-2. Investigations have not to date delineated the reason for these unusual retention patterns. These investigations are continuing.

Conclusions

The following basic conclusions were reached as a result of this study.

(1) There are certain forms of uranium handled in the Y-12 Plant which have long biological half-lives in the human chest.

(2) The chances of significant exposure to

such materials under the work conditions experienced in the Y-12 Plant over the last 4 years are relatively remote.

(3) Fecal excretion is a significant mode of elimination in such cases.

(4) High and variable chest-burden-to-excretion-rate ratio make the detection and evaluation of such cases by excreta analyses alone extremely difficult if not impossible.

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