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ESTIMATION OF ^{244}Cm INTAKE BY BIOASSAY MEASUREMENTS FOLLOWING A CONTAMINATION INCIDENT*

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ABSTRACT

An employee was contaminated with radioactive material consisting primarily of ^{244}Cm and ^{246}Cm as a consequence of handling a curium nitrate solution at a reprocessing facility. *In vivo* gamma analysis and *in vitro* (urine and fecal) analysis were initiated soon after the incident. Further *in vivo* measurements were performed regularly through hour 528, and *in vitro* bioassay measurements were obtained through day 14. A sample of the curium solution from the workplace was obtained to confirm that the nitrate was the chemical form and to identify the curium isotopes present. The mass ratio of ^{244}Cm:^{246}Cm was determined to be 91:7. Diethylenetriaminepentaacetate (DTPA) was administered on hours 33 and 71. Observed excretion rates were consistent with available information for curium in the literature. In this paper, the results of the *in vivo* and *in vitro* measurements are presented and intake estimates for the incident are developed using various excretion rate functions.

INTRODUCTION

An Oak Ridge National Laboratory employee was contaminated with ^{244}Cm in an incident at a reprocessing facility where curium was being handled. The radioactive material involved was in the form of curium nitrate solution containing ^{244}Cm and ^{246}Cm. Mass spectrometric determination[1] showed that the material contained 91% ^{244}Cm and 7% ^{246}Cm by mass, corresponding to an activity ratio of 3400:1. The remaining mass fraction included trace levels of ^{243}Cm and ^{239}Pu. It was thus concluded that ^{244}Cm was the nuclide of radiological concern. The

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radiological characteristics[2,3] of the curium radionuclides involved in this incident are given in Table 1.

It is believed that the incident occurred as the employee attempted to dilute a curium nitrate solution in a chemical hood. Contamination of the hood was discovered and reported, and, upon further investigation, it was determined that the incident probably occurred late in the previous workshift. An air monitor located in the laboratory indicated no abnormal airborne activity during the period and nasal smears of two workers indicated no detectable activity. All workers involved were counted at the ORNL Whole-Body Counting Laboratory and urine samples were collected for analysis.

Only limited information is available on the metabolism of internally deposited ^{244}Cm. Information on the excretion and retention of curium is needed to estimate a worker's intake from bioassay measurements and to estimate the radiation dose associated with the intake. The International Commission on Radiological Protection (ICRP) in its recent review[4] of the metabolism of plutonium and related elements suggests that 90% of the curium reaching the blood stream will deposit equally between the liver and bone where it will be retained with halftimes of 20 and 50 years, respectively. The Commission also recommended a fractional absorption (f_1) from the gastrointestinal tract to blood of 10^{-3} for curium.[4] The review does not provide any guidance regarding the urinary excretion of curium. Information that is available indicates that ^{244}Cm behavior in the body contrasts sharply with that of plutonium, and may be more like americium. Sanders[5], Parkinson *et al.*[6] and Cohen *et al.*[7] have examined the excretion of curium in man and in the baboon in attempts to define the excretion functions.

In the present paper, we apply available curium excretion-rate functions to an actual exposure incident in an attempt to estimate the worker's curium intake.

IN-VIVO MEASUREMENTS

The first in vivo bioassay (whole-body counting) was performed approximately 20 hours after the incident and continued on a regular basis through hour 528. Whole body counting is performed at ORNL following any potential exposure to actinides. The radiations of interest for ^{244}Cm (see Table 1) are the internal conversion L x-rays.

The major contributor to the background in the L x-ray region is Compton scatter from ^{40}K, which occurs naturally in the human body. This contribution was estimated, for this subject, from measurements of

50 adult male non-radiation workers. Calibration factors, adjusted for tissue thickness,[8] were obtained from an anthropomorphic phantom with ^{244}Cm-loaded lungs and liver. The calibration factor used for pulmonary deposition of ^{244}Cm was 600 Bq/cpm,* based on an exponentially averaged chest-wall thickness of 3.26 cm.

Standard counting protocol was used during all whole body and organ counts.[9] Count times were 1600 seconds for the lungs and 1000 seconds for the liver and skull using a 5-in.-diam. phoswich detector system. The results of the lung counting are shown in Fig. 1.

IN-VITRO MEASUREMENTS

In-vitro bioassay measurements for ^{244}Cm in urine and feces began at about hour 32 after the incident. Fecal analysis was continued to day 46 and urinalysis continued until day 74. The urine and fecal samples were prepared and radiochemical analysis performed by standard procedures[10]. A total of 26 urine and 15 fecal samples were analyzed. Alpha spectrometry (silicon surface barrier detector) was used to identify and quantify curium activities in each sample. Each sample was counted for at least 1000 minutes to obtain a one-sigma propagated error of less than 20%. The results of ^{244}Cm in urine and in feces as a function of time are shown in Fig. 2 and Fig. 3, respectively.

INTERPRETATION OF DATA

It can be seen from the results of lung counts shown in Fig. 1 that the amount of radioactivity decreased significantly during the first few analyses performed on the first day after the incident. Activity assigned to the thorax was reduced 85% by removing the subject's beard. This suggests strongly that the beard was contaminated, and that the initial lung counts were influenced by this external contamination. There was no indication from an operating air monitor that activity became airborne in the work area where the incident occurred. Nasal swabs and whole body counts from two co-workers indicated no elevated alpha activity. The minimum detectable activity for ^{244}Cm at the 99% confidence interval in the lung of this individual was 470 Bq; the lung count results were not statistically different from those expected for an uncontaminated adult male by day 4. The lack of evidence for airborne radioactivity, uncontaminated nasal swabs, and lung count results that seem to indicate surface, rather that internal, contamination, combine to suggest that the initial intake was not by

* cpm = counts per minute

inhalation. We assume that ingestion was the principal intake mode. This assumption is also supported by a relatively large fecal excretion of ^{244}Cm on the third day.

Fig. 2 gives a plot of the urinalysis results during the first 75 days. Different functional forms were tried. We finally decided on the form $a_1 e^{-b_1 t} + a_2 e^{-b_2 t}$ + constant. The constant was set equal to 0.0018, which is the minimium of the data points. Then a_1, a_2, b_1, and b_2 were found by least square fit. The least square fit yielded

$$Y_u(t) = 0.2253\ e^{-0.5t} + 0.00403\ e^{-0.05t} + 0.0018 \quad , \qquad (1)$$

where $Y_u(t)$ is the urinary excretion rate in Bq/d. The infinite-time integral of the first two terms of Eq. 1 gives an estimate of 0.53 Bq involved in this excretion route. DTPA administration had no apparent effect on the excretion pattern.

Results of the fecal analyses are shown in Fig. 3. We assumed that the excretion data could be described by the functional form $a\ e^{-b\ t}$. A least square fit to the fecal excretion data gave the regression line

$$Y_f(t) = 3.71\ e^{-0.2t} \quad , \qquad (2)$$

where $Y_f(t)$ is the fecal excretion rate in Bq/d. The constant b corresponds to a biological halftime of about 3.5 days. No attempt was made to include the first datum in the regression analysis since it is probably associated with clearance of unabsorbed curium rather than biliary excretion of systemic curium. The total activity measured in all fecal samples collected was 5.7 Bq.

Parkinson *et al.*[6] developed a urinary excretion function based on the rate of excretion of ^{244}Cm by two workers who had sustained accidental uptakes through a puncture wound and an acid burn of the skin. An excretion function $E_u(t)$ was derived as

$$E_u(t) = 0.053\ e^{-0.935t} + 0.0019\ e^{-0.0733t} + 0.00016\ e^{-0.00899t} + 8.6 \times 10^{-6}\ e^{-0.0000190t} \quad , \qquad (3)$$

where $E_u(t)$ is the fraction of the uptake at t=0 excreted per day at time t.

Sanders[5] described the excretion of curium in urine and feces of two workers who had inhaled curium. One case (Sanders' case I) involved inhalation of "relatively soluble" compounds of ^{244}Cm. From the details given by Sanders[5] one can derive a urinary excretion function by

assuming that curium was instantaneously transferred from the lung to blood (i.e., ignoring lung clearance kinetics). The resulting urinary excretion function is

$$E_u(t) = 0.051\ e^{-0.067t} + 0.00011\ e^{-0.0089t} \quad . \tag{4}$$

If one assumes that curium's behavior in the body is similar to americium, use can be made of Rosen's americium excretion function[11] for adult baboons. The urinary excretion rate following an intravenous injection of activity in the baboon is

$$E_u(t) = 0.036t^{-1.3} \quad , \tag{5}$$

for t>1 day.

The intake <I> by ingestion can be estimated as

$$\langle I \rangle = \frac{1}{n\ f_1} \sum_{i=1}^{n} \frac{Y_u(t_i)}{E_u(t_i)} \quad , \tag{6}$$

where $Y_u(t_i)$ denotes the urinary excretion rate observed in the ith sample obtained at time t_i in Bq/d,

f_1 is the fractional transfer of curium from the gastrointestinal tract to blood (a value of 10^{-3} has been used),

$E_u(t_i)$ is the expected urinary excretion rate at time t_i per unit activity reaching blood (1/d), and

n is the total number of urine samples.

Estimates of ^{244}Cm intake for the incident are provided in Table 2. They are derived from the excretion function of Parkinson, *et al.*[6], a function derived from Sanders[5], and application of Rosen's Am excretion function.[11]

DISCUSSION

Contamination discovered in a chemical laboratory apparently occurred as an employee attempted to dilute a curium nitrate solution. *In-vivo* and *in vitro* analysis were begun to estimate the magnitude of the worker's intake which appeared to be by ingestion. The chemical form and isotopic composition of the contamination was determined by analyzing a sample of the solution handled by the employee. *In-vivo* and *in vitro* analysis were begun as soon as possible and estimates of the apparent intake were derived from the excretion using urinary excretion models form the literature.

Estimates of the amount of material ingested ranged from 0.5 to 8 kBq or about 1 to 16% of the Annual Limit on Intake (ALI)[2] for ^{244}Cm tabulated in Table 1. Using the committed effective dose equivalent per unit intake shown in Table 1, the intake estimates correspond to a committed effective dose equivalent ranging from 0.27 to 4.3 mSv. The variations in the estimated intake arise from the approximations being made (e.g., assumption of instantaneous uptake to blood, and the nature of the urinary excretion function used in the analysis). Despite the limitations inherent in estimating intake from excretion data, this often remains the best approach for quantifying internal exposures to curium and other transuranic alpha-emitters with elevated MDAs *in vivo*. The range of intake estimates presented here indicates the need for additional research on curium metabolism, not only to provide the basis for establishing radiation protection guidance, but also to provide the means of evaluating the intakes that may result from handling curium in the workplace.

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DISCLAIMER

Table 1.

Pertinent Radiological Characteristics of Curium Radionuclides

Cm-243 $T_{\frac{1}{2}}$ = 28.5 y — Principal Radiations Emitted

Radiations	Energy (keV)	Intensity (%)
Alpha	5742	11.5
	5785	73.3
$K\alpha_1$ x-ray	103.8	22.9
$K\alpha_2$ x-ray	99.6	14.3
$L\alpha$ x-ray	14.3	23.4

Radiation Protection Guidance*

Intake Mode	ALI (kBq)	He (Sv/Bq)**
Inhalation	0.3	8.0×10^{-5}
Oral	40	5.7×10^{-7}

Cm-244 $T_{\frac{1}{2}}$ = 18.11 y — Principal Radiations Emitted

Radiations	Energy (keV)	Intensity (%)
Alpha	5763	23.6
	5805	76.4
$L\alpha$ x-ray	14.3	3.77
$L\beta$ x-ray	18.1	4.83

Radiation Protection Guidance

Intake Mode	ALI(kBq)	He(Sv/Bq)
Inhalation	0.4	6.4×10^{-5}
Oral	50	5.4×10^{-7}

Cm-246 $T_{\frac{1}{2}}$ = 4730 y — Principal Radiations Emitted

Radiations	Energy (keV)	Intensity (%)
Alpha	5343	21.0
	5387	79.0
$L\alpha$ x-ray	14.3	3.34
$L\beta$ x-ray	18.1	4.31

Radiation Protection Guidance

Intake Mode	ALI(kBq)	He(Sv/Bq)
Inhalation	0.2	1.2×10^{-4}
Oral	30	1.0×10^{-6}

*The Annual Limit on Intake (ALI) for the curium isotopes has been computed using the metabolic information in ICRP-[illegible]. For inhalation curium compounds are assigned to lung clearance class W and an f_1 value of 0.001 was assumed.

**He denotes the effective committed dose equivalent per unit intake by inhalation or oral intake.

Table 2.

Estimated intake of ^{244}Cm

Excretion Function	Intake (kBq)
Parkinson (Eq. 3)	8
Sanders (Eq. 4)	0.5
Rosen (Eq. 5)	5

Fig. 1. Measured ^{244}Cm activity in chest as a function of time. For this subject the MDA was 470 Bq.

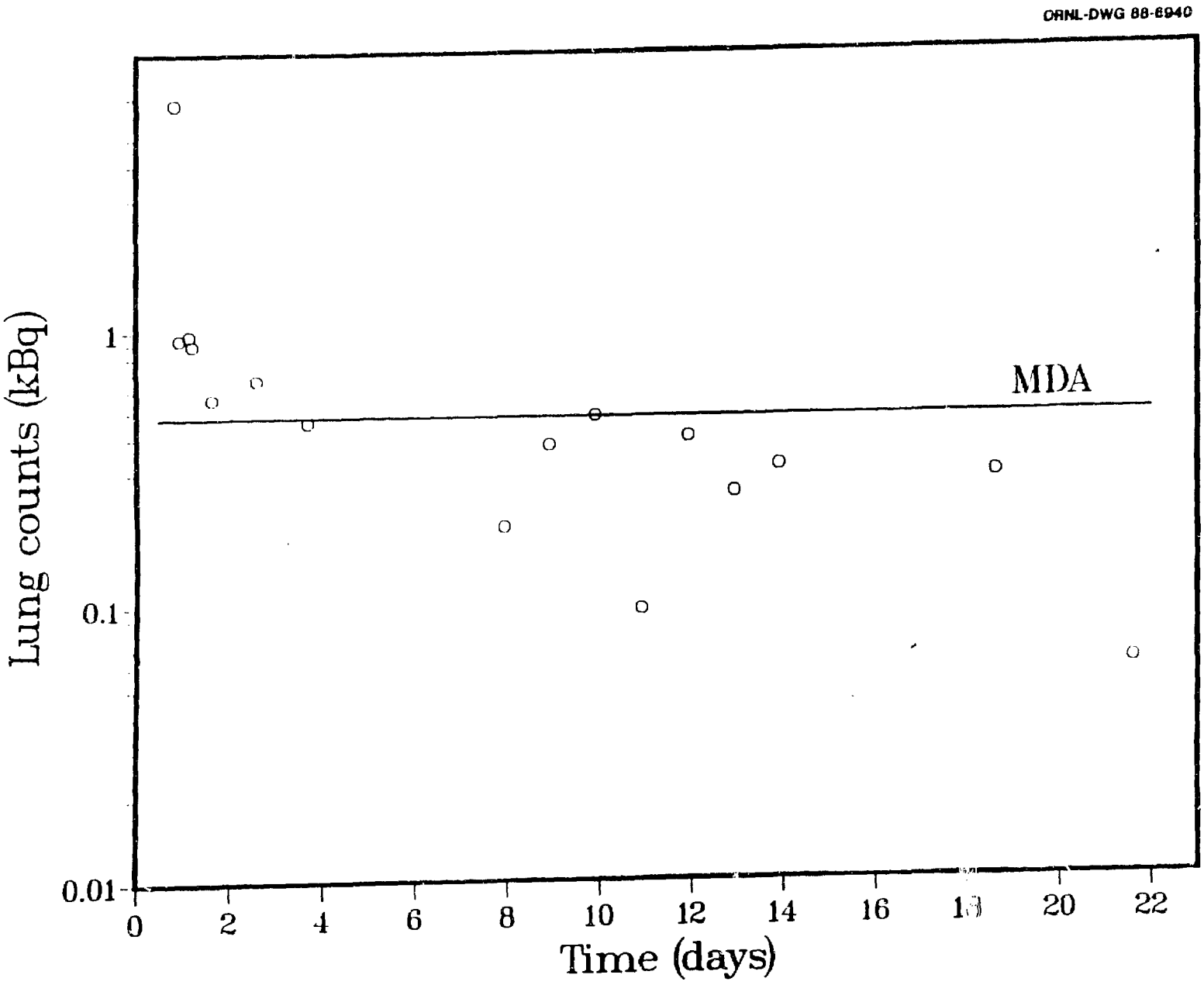

ORNL-DWG 88-6940
Lung counts (kBq)
1
0.1
0.01
MDA
0
2
4
6
8
10
12
14
16
18
20
22
Time (days)

Fig. 2. Urinary elimination rate of ^{244}Cm.

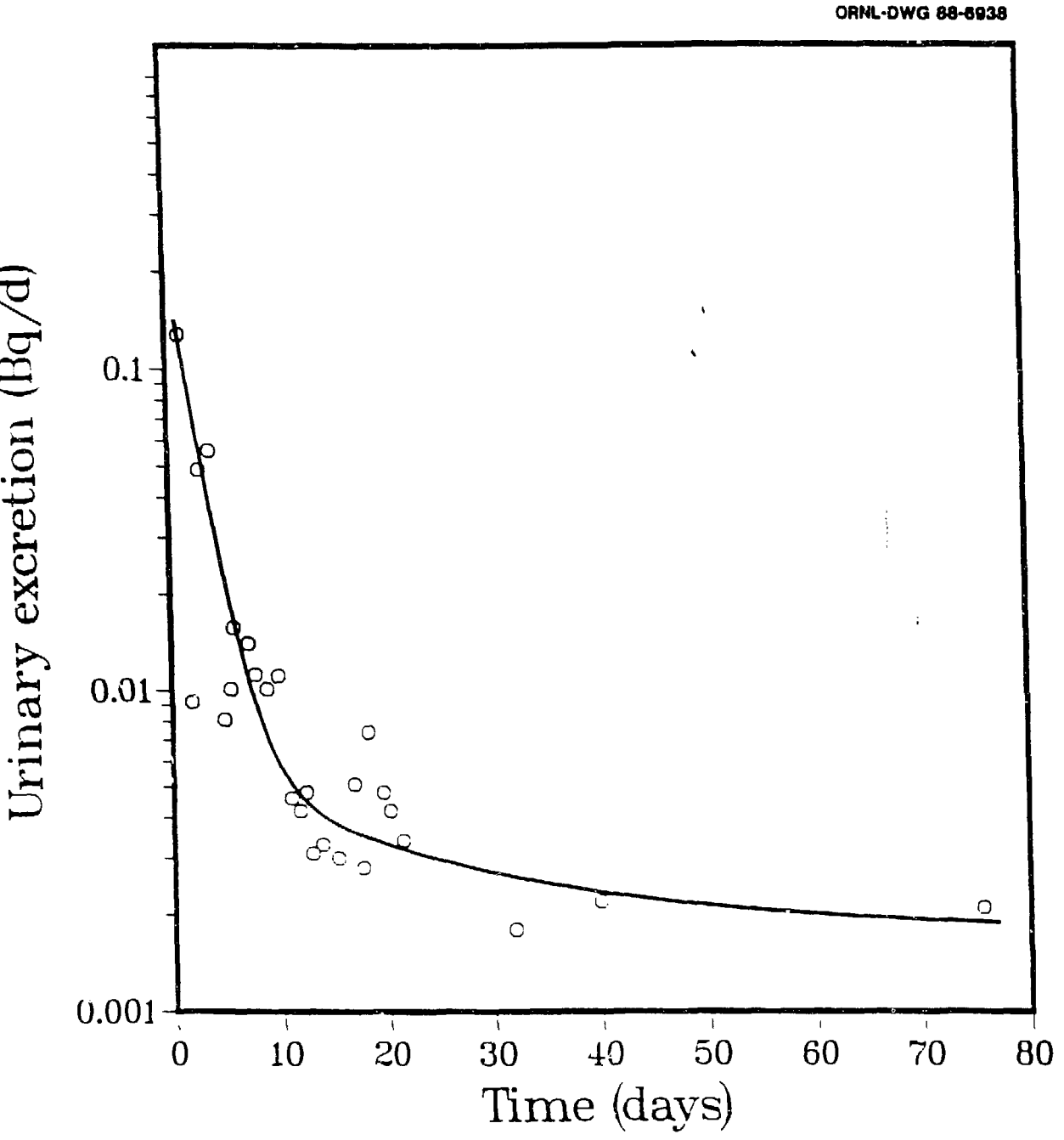
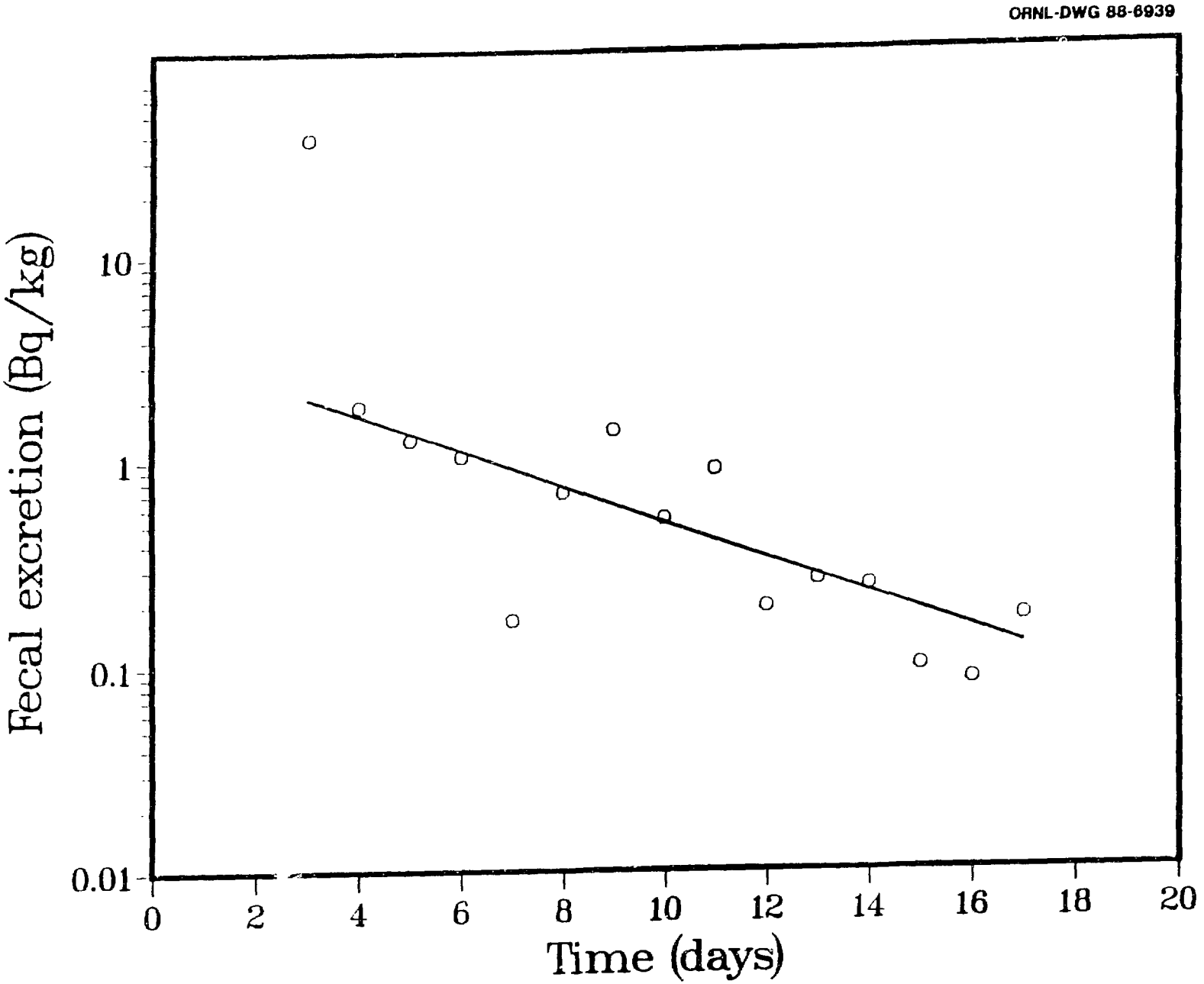

ORNL-DWG 88-6938
Urinary excretion (Bq/d)
0.1
0.01
0.001
0
10
20
30
40
50
60
70
80
Time (days)

Fig. 3. Fecal elimination rate of ^{244}Cm.


ORNL-DWG 88-6939
Fecal excretion (Bq/kg)
10
1
0.1
0.01
0
2
4
6
8
10
12
14
16
18
20
Time (days)