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ESTIMATION OF DOSE COMMITMENT FROM AN ACCIDENTAL INTAKE OF ^{244}Cm*

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Four employees at the Oak Ridge National Laboratory experienced an intake by inhalation of ^{244}Cm as a result of an accidental release of this radionuclide. Urine and fecal samples were obtained and were analyzed in the Bioassay Laboratory at ORNL. Also, chest counts were made with the ORNL Whole Body Counter. Ca-DTPA mists were inhaled within hours after the intake. These data together with the data of McClellan et al. (Health Physics Journal, 27, 359, 1974) on the inhalation of ^{244}Cm in the dog were used to obtain a 50-year dose commitment to lungs, skeleton, and liver. The 50-year dose commitment to skeleton for two of the employees is estimated to be about 170 rem. The estimation methods used for dose commitment together with the values for other organs obtained are presented and discussed.

I. INTRODUCTION

Four employees of ORNL took into their bodies by inhalation ^{244}Cm which was accidentally leaked into the breathing zone of these workers. Chest counts and sputum samples were made and urine and fecal samples were collected and the latter were analyzed in the

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bioassay laboratory. The employees were also given Ca-DTPA, by inhalation, shortly after the intake.

The purpose of this study was the estimation of the one-year and 50-year dose commitment to those organs and tissues which take up the ^{244}Cm. In the following, the data on the human subjects and some data on dogs reported by McClellan et al. (1972) will be discussed and estimations of the dose will be presented. Also, a discussion of the estimates of the effect of the Ca-DTPA on elimination will be given.

II. DATA AND COLLECTION

A. Human data

Table 1 presents a summary of the data on urine, feces, sputum, and chest counts for the four employees. In the first column appears the identification of the employee, the second column is the day post-intake of the ^{244}Cm, the third column lists the chest count in nCi, the fourth lists the activity (d/m/da) excreted in feces, the fifth the activity (d/m/24 hours) excreted in urine and the sixth lists the activity (d/m) present in sputum. Also shown in column 4 is the total activity (d/m and nCi) excreted in the feces of these employees in the first one or two weeks.

B. Tissue data and excretion data on experimental dogs. (Data of McClellan et al.)

Urinary excretion data on dogs are shown in Figure 1. The data are plotted in percent of the initial lung burden per day versus the days post-intake by the dogs. These data show that, on the first day, about 5% per day of the initial lung burden is excreted in urine. Thus, if the initial lung burden is known, the approximate urinary excretion can be estimated.

The burden in the lungs, skeleton, and liver of dogs after inhalation of ^{244}Cm as the chloride and the oxide are shown in Figure 2. These data are presented in percent of initial lung burden vs. the time post-inhalation exposure, in days. As can be seen, the burden decreases in lungs and increases in skeleton and liver and then levels off or decreases slightly. Note that the skeleton takes up a maximum of about 40 to 50% of the initial lung burden while liver takes up a maximum of around 30% of the initial lung burden and then decreases, slightly, with time.

C. Human chestcount data

The activity (nCi) detected in the lungs (chest) of two of the workers as a function of the time after the accident is shown in Figure 3. Also shown is a curve fitting, approximately, the data on retention in the lung of the dog. The chest counts of Employee B decrease with time in a manner that is approximately parallel to the data on the dog. From this Figure it appears that the initial lung burden of the Employees A and B is about 20 nCi. Some points are plotted in the margin of the graph because they are time zero points.

D. Calculated excretion rates of the employees from the chest burden data and the dog inhalation data

The urinary excretion data on the dogs in Figure 1, show that approximately one-twentieth of the initial lung burden is excreted on the first day. Further, we have estimated the initial lung burden in two of the employees to be $\sim$20 nCi. We The estimated excretion as a function of time plotted with the measured values appear in Figure 4.

The data on all employees (the values in column 5 of Table 1) are plotted together with the estimated excretion values on days 1, 4, and 8. As can be seen, there is general agreement between the estimated and measured urinary excretion of ^{244}Cm.

E. Estimated one-year and 50-year dose commitment for the four employees

In order to estimate dose commitments for this exposure, decay scheme data for ^{244}Cm is needed. These data are presented in Table 2 (Dillman, 197). Column 1 gives the type of radiation emitted. Column 2 gives the abundance of the emitted radiation in units of percent per decay. The energy (in MeV) of the radiation is listed in column 3. About 3/4 of the emissions are $\sim$ 5.8 MeV alpha particles while the other 1/4 is $\sim$ 5.7 MeV alpha particle. There are also gamma rays, x-rays, and conversion and Auger electrons; however, these do not contribute significantly to the total energy emitted when compared with the alpha particle energy.

For dose to the skeleton we assume an alpha particle energy of 5.7 MeV, a quality factor and a modifying factor of 50 (10 for Q and 5 for factor required by ICRP Committee 2 in its Publication 2 report) and 7000 grams of bone (the skeleton weighs 10,000 grams but this includes 3000 grams of marrow--we only use the bone weight) and a radiological half-life of 6500 days (from Table 2). Substituting into the equation

$$\text{Dose} = \frac{51.15 \times E \times}{M} \int_0^{50\ \text{yrs}} q e^{-\lambda_r \tau}\, d\tau \text{ rems}$$

Where E is the absorbed energy in MeV, M is the mass of tissue in grams and the integral is the residence time or cumulated activity (μCi-days) in the organ. For liver and skeleton we assume no biological elimination and an initial burden of 6 nCi and 10 nCi, respectively. For lung, we assume the power function retention pattern found in dogs and an initial burden of 20 nCi. Thus, our estimated 50-year dose commitments are 80 rem, 170 rem, and 7 rem for the liver, skeleton, and the lung, respectively. In the estimates of dose to the lung we have neglected the radiological decay of ^{244}Cm.

An estimate of 1-year dose commitment for skeleton was about 7 rem, which is nearly 1/4 the permissible yearly bone dose limit of 30 rem.

III. DISCUSSION AND CONCLUSIONS

The effect of Ca-DTPA administered the employees by inhalation shortly after the intake should be discussed. It appears that the only effect occurs 7 or 8 days after the intake where the excretion is somewhat elevated over the data for the dog (see Figure 4). This elevated excretion at one to two weeks may be due to a species effect--urinary elimination by man may be more rapid than urinary excretion by dog of inhaled $^{244}CmCl_3$. However, this is usually not the case in most of the studies of excretion of injected radionuclides as observed by Richmond et al. (Richmond, 1958). In their studies (not on Cm but on alkali metals, Se, Mn, Co, etc.) the excretion from the body is generally greater as one goes from man through dog, monkey, rat and down to mouse. More data are needed on Cm metabolism in these other species.

In conclusion it appears Employees A and B received a 50-year dose commitment to bone of about 170 rem while doses to bone of Employees C and D were about 1/500 of this level or about 400 mrem. (See Figure 4 for urinary excretion where C and D are about 1/500 the level of A and B.) The 50-year dose commitment to liver of Employees A and B was about 70 rems while for C and D it is about 150 mrem. Dose commitments to lung (50-year) are less than 7 rem for A and B and less than 15 mrem for C and D.

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Table 1

Excretion and Lung Burden Data on the Four Employees

Employee	Day	Lung Count nCi	Feces d/m per day	Urine d/m per 24 hrs	Sputum d/m per specimen
A	0	22			700
	1	13	3.1E4	239	
	2		8.6E4	1189	
	3	10	1.8E4	864	6
	4	MD	7.3E2	291	
	5		5.1E2	374	
	6		2.5E2	420	
	7			484	
	8	MD		384	
	9		3.8E2	312	
	10	MD			
	14	MD			
			1.37×10^5 (62 nCi)		
B	0	27			74
	1	20	4.7E2	5	
	2		4.2E3	230	
	3	18	2.1E3	147	2
	4	20		118	
	5		1.6E2	118	
	7		5.0E0	172	
	8	10		110	
	9			91	
	10	8		92	
	14	4±2			
			6.9×10^3 (3.1 nCi)		
C	0	MD			8
	1		4.0E1	3	
	2		3.7E2	8	
	3	MD	5.0E1	5	
	4		5.0E0	5	0
	5			4	
	7			2	
			4.65×10^2 (.21 nCi)		

Table 1 (cont'd)

Employee	Day	Lung Count nCi	Feces d/m per day	Urine d/m per 24 hrs	Sputum d/m per specimen
D	0	MD			24
	1			10	
	2		1.9E2	6	
	3	MD	1.3E2	12	
	4		6.0E1	6	0
	7			7	
			3.8×10^2 (.17 nCi)		

MD: At or below minimum detectable of 5 ± 4 nCi.

Table 2

Decay Scheme Data for ^{244}Cm
(Halflife = 1.785E 01 Years)
(Mode of Decay, Alpha)

Type of radiation	Percent per decay	Energy (MeV)*
Alpha 1	0.0230	5.66660
Recoil Atom	0.0230	0.09444
Alpha 2	23.2920	5.76380
Recoil Atom	23.2920	0.09606
Alpha 3	76.6850	5.80600
Recoil Atom	76.6850	0.09677
Gamma Ray 1	10.3987	0.04288
L Shell Conversion Electron	9.6875	0.02133
M Shell Conversion Electron	3.2288	0.03832
Gamma Ray 2	0.0202	0.09882
L Shell Conversion Electron	0.0021	0.07727
L-Alpha X-Ray	2.1229	0.01426
L-Beta X-Ray	1.9890	0.01795
L-Gamma X-Ray	0.2678	0.02142
LMM Auger Electron	5.3099	0.01244
MXY Auger Electron	18.2290	0.00456

*Average energy for beta plus or beta minus.

Hamilton having reported less than 0.05% gastrointestinal absorption for ^{242}Cm in rats.[11] This aspect of ^{244}Cm metabolism has apparently not been studied extensively and is deserving of additional attention. With both forms, ^{244}Cm which did not clear rapidly was avidly retained with a mean retention half-time of 580 days (range of 250–1400 days). Recognizing that this half-time was obtained from data collected over a relatively short time period, confirmation with longer term observations will be necessary to arrive at more definite estimates.

Urinary excretion patterns of ^{244}Cm following inhalation of the two forms of ^{244}Cm (Fig. 2) show a striking similarity. This suggests that the two aerosol forms were being solubilized in the lung, entered the blood stream, and were available for excretion via the kidneys in a similar fashion.

This striking similarity of dissolution and clearance of the two different aerosols from the lung is further borne out by the distribution of activity in three major tissues: lung, liver and skeleton (Fig. 3). With both forms, the ^{244}Cm rapidly left the lung such that by 16 days post-inhalation exposure, less than 20% of the initial lung burden of ^{244}Cm remained in lung. Curium-244 that left the lung was primarily deposited in skeleton and liver. Lesser amounts of ^{244}Cm were deposited in other tissues, as shown in Table 2. The tissue concentrations shown have been normalized to a 10-kg dog and with only one exception, were quite similar at any given time for dogs exposed to the two forms. The exception was in the turbinate samples, which were higher in ^{244}Cm concentration in the $^{244}CmCl_3$ dogs. This suggests a greater deposition and retention of ^{244}Cm in the turbinates of these dogs exposed to the aerosols with AMAD ~1.6 μm as compared to the $^{244}CmO_{1.73}$ dogs exposed to aerosols with AMAD ~0.5 μm. Of interest was the relatively high concentrations of ^{244}Cm in the thyroid, especially at early times when it exceeded the concentration of ^{244}Cm found in liver or skeleton. This finding is similar to that of LLOYD *et al.*[13] who noted thyroid concentrations of ^{241}Am about a factor of 2 less than liver concentrations for dogs given ^{241}Am citrate intravenously.

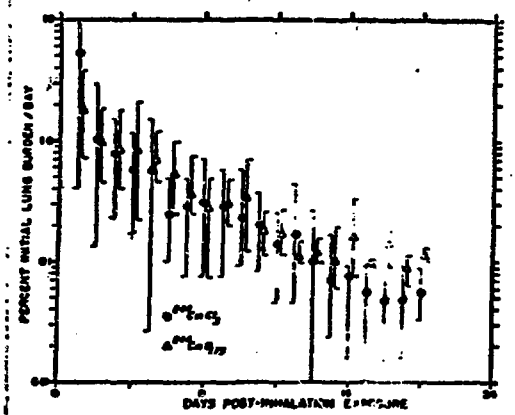

FIG. 2. Urinary excretion of inhaled ^{244}Cm in Beagle dogs. Datum points are the mean values, vertical brackets represent the range.

The similarity of the tissue distributions of ^{244}Cm following inhalation of the two chemical forms suggests that in both cases the material leaving the lung was probably in the same form. The relative solubility of the $^{244}CmCl_3$–CsCl is not unexpected considering that the ^{244}Cm represented only a small fraction (≈0.00057) by mass of the aerosol particles. Thus, predicting that the CsCl was rapidly absorbed[14] only a small amount of ^{244}Cm (≈0.00057) of the original particle by mass, would be left. Presumably this small amount could be rapidly complexed in a form that would favor rapid translocation. The extent to which larger respirable particles of pure $^{244}CmCl_3$ would behave in like fashion is not known and is deserving of investigation. More mass might favor the formation of more insoluble complexes resulting in higher lung retention with less translocation to liver and skeleton. Such a mass or carrier effect has been seen with inhaled radioyttrium.[15] On the other hand, the inherent solubility of ^{244}Cm and the complexes it might form with organic molecules might be such that additional curium per particle might have little effect on lung retention and translocation to other tissues.

Because oxides of the actinides are generally accepted as being relatively insoluble,[12,16] one might have expected that the $^{244}CmO_{1.73}$ aerosol would be more avidly retained in lung. Such, however, was not the case. This may be related to the inherent solubility of $^{244}CmO_{1.73}$

Fig 2

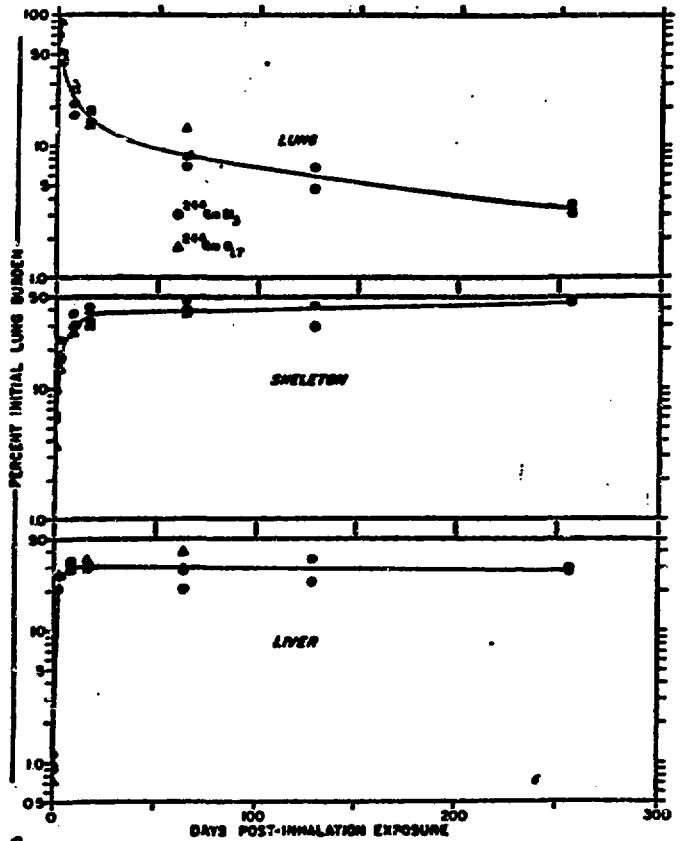

FIG. 2. Lung, liver and skeletal retention of inhaled ^{244}Cm in Beagle dogs. Datum points are the actual values for individual animals. The smooth curves were hand fit by eye to the datum points.

and/or the relatively small size of the particles inhaled, recognizing that the dissolution of particles is related to their surface area (and the ratio of surface area to mass or activity is high for small particles).[17] The present data do not rule out the possibility of the rapid dissolution being related to particle size since the mass of ^{244}Cm present in the $^{244}CmO_{1.73}$ particles was on the average only about 5 times that present in the $^{244}CmCl_3$–CsCl particles. (This is based on the difference in the mass, density and chemical composition of the aerosols. The median real diameter of the $^{244}CmO_{1.73}$ and $^{244}CmCl_3$–CsCl aerosols was 0.03 μm versus 0.8 μm, respectively, for a factor of ~1000 difference in volume, resulting in an ~300-fold difference in mass when the densities of 10.5 and 3.2, respectively, are taken into account. This 300-fold difference is narrowed when it is recognized that 0.00057 of the $^{244}CmCl_3$–CsCl aerosol was ^{244}Cm compared to 0.9 of the $^{244}CmO_{1.73}$ aerosol.)

The extent to which the high solubility observed may be related to the inherent solubility of the $^{244}CmO_{1.73}$ deserves additional study; in particular, data should be obtained on the solubility of inhaled $^{244}CmO_{1.73}$ particles of

Fig 3

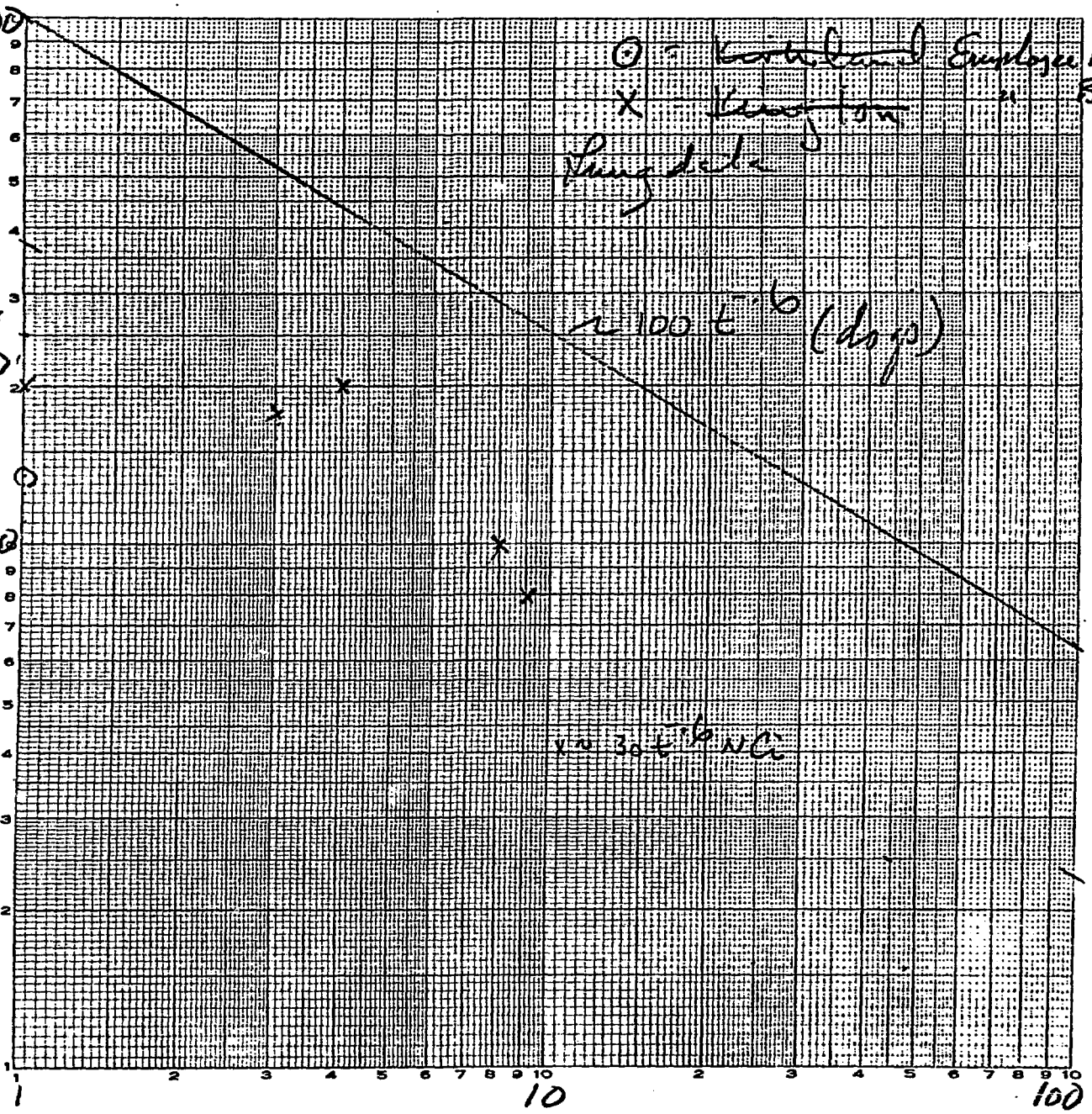
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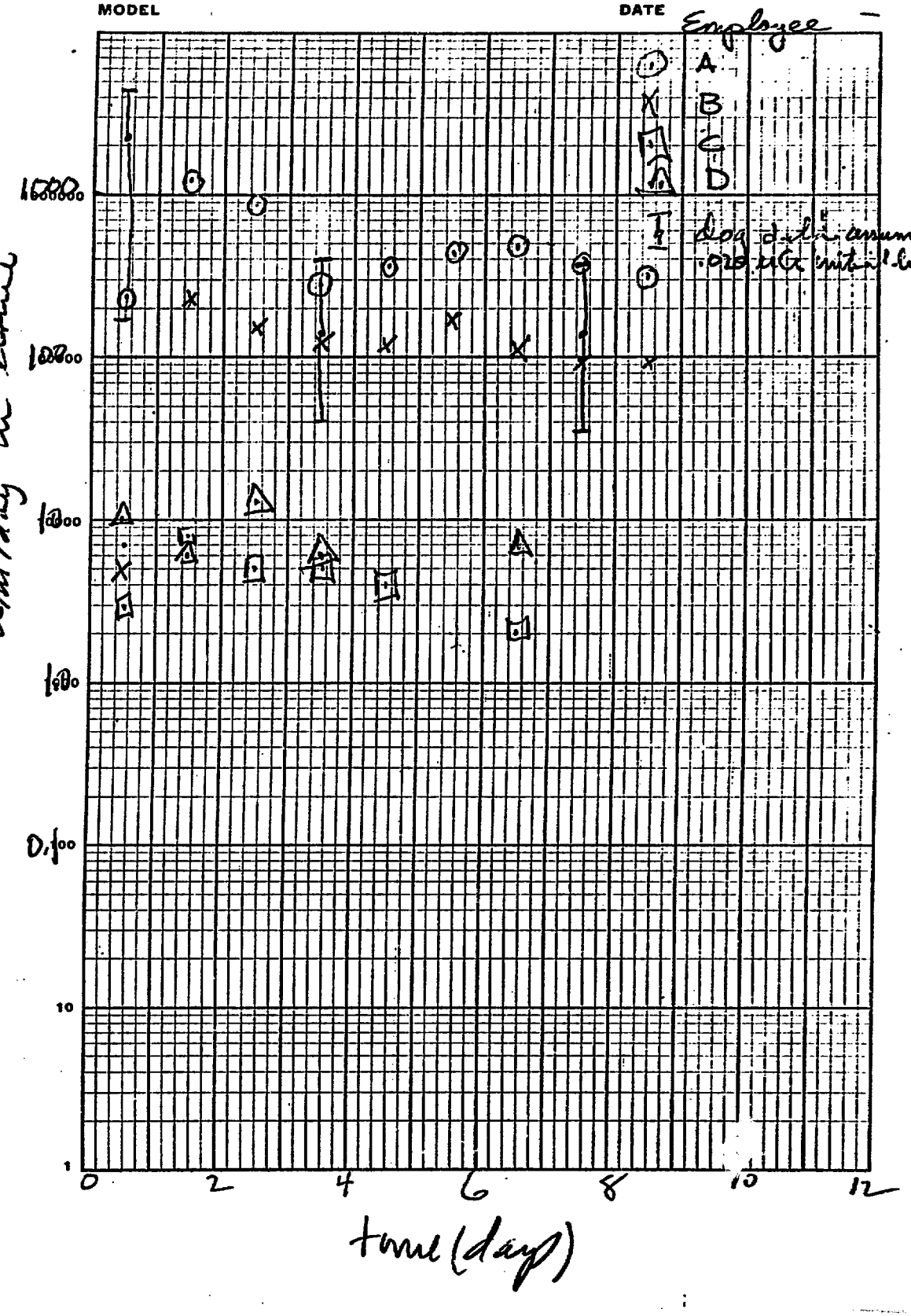
Fig. 4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